



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US99/05285 <b>(22) International Filing Date:</b> 10 March 1999 (10.03.99) <b>(30) Priority Data:</b> 60/077,534                      11 March 1998 (11.03.98)                      US <b>(71) Applicant (for all designated States except US):</b> THE DOW CHEMICAL COMPANY [US/US]; 2030 Dow Center, Midland, MI 48674 (US). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> TURLEY, Robert, R. [US/US]; 606 Lake Road, Lake Jackson, TX 77566 (US). STEWART, Kenneth, B. [US/US]; 107 Marigold, Lake Jackson, TX 77566 (US). <b>(74) Agent:</b> SPENCER, Lee; The Dow Chemical Company, Patent Dept., B-1211, 2301 Brazosport Boulevard, Freeport, TX 77541 (US).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> FIBERS MADE FROM $\alpha$ -OLEFIN/VINYL OR VINYLIDENE AROMATIC AND/OR HINDERED CYCLOALIPHATIC OR ALIPHATIC VINYL OR VINYLIDENE INTERPOLYMERS		
<b>(57) Abstract</b>  <p>The present invention pertains to fibers comprising: (A) from 50 to 100 wt. percent (based on the combined weights of Components A and B) of at least one substantially random interpolymer having an <math>I_2</math> of from 0.1 to 1,000 g/10 min, a density greater than 0.9300 g/cm<sup>3</sup>, and an <math>M_w/M_n</math> of 1.5 to 20; which comprises: (1) from 0.5 to 65 mol percent of polymer units derived from: (i) at least one vinyl or vinylidene aromatic monomer, or (ii) at least one hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or (iii) a combination of at least one aromatic vinyl or vinylidene monomer and at least one hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and (2) from 35 to 99.5 mol percent of polymer units derived from ethylene or at least one C<sub>3-20</sub> <math>\alpha</math>-olefin or a combination thereof; and (B) from 0 to 50 percent by weight (based on the combined weights of Components A and B) of at least one takifier. The fibers of the present invention could have applications such as carpet fibers, elastic fibers, doll hair, personal/feminine hygiene applications, diapers, athletic sportswear, wrinkle free and form-fitting apparel, conductive fibers, upholstery, and medical applications including, but not restricted to, bandages, gamma sterilizable non-woven fibers.</p>		

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FIBERS MADE FROM ALPHA-OLEFIN/VINYL OR VINYLIDENE AROMATIC  
AND/OR HINDERED CYCLOALIPHATIC OR ALIPHATIC VINYL OR  
VINYLIDENE INTERPOLYMERS

5           This invention is related to fibers and to fabrics and articles fabricated therefrom. The fibers are prepared from polymers which comprise at least one substantially random interpolymer comprising polymer units derived from one or more  $\alpha$ -olefin monomers with one or more vinyl or vinylidene aromatic monomers and/or hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers.

10           A variety of fibers and fabrics have been made from thermoplastics, such as polypropylene, highly branched low density polyethylene (LDPE) made typically in a high pressure polymerization process, linear heterogeneously branched polyethylene (for example, linear low density polyethylene made using Ziegler catalysis), blends of polypropylene and linear heterogeneously branched polyethylene, blends of linear  
15 heterogeneously branched polyethylene, and ethylene/vinyl alcohol copolymers.

          Of the various polymers known to be extrudable into fiber, highly branched LDPE has not been successfully melt spun into fine denier fiber. Linear heterogeneously branched polyethylene has been made into monofilament, as described in USP 4,076,698 (Anderson et al), and into fine denier fiber, as disclosed in USP  
20 4,644,045 (Fowells), USP 4,830,907 (Sawyer et al.), USP 4,909,975 (Sawyer et al.) and in USP 4,578,414 (Sawyer et al.).

          Blends of such heterogeneously branched polyethylene have also been successfully made into fine denier fiber and fabrics, as disclosed in USP 4,842,922 (Krupp et al.), USP 4,990,204 (Krupp et al.) and USP 5,112,686 (Krupp et al.).

25           In addition to heterogeneously branched LLDPE, fibers have also been made from narrow molecular weight distribution ethylene copolymers produced using the so called single site catalysts as described by Davey et al., in USP 5,322,728 and WO 94/12699.

          Fibers have also been made from other polymeric materials. USP 4,425,393  
30 (Benedyk) discloses monofilament fiber made from polymeric material having an

elastic modulus from 2,000 to 10,000 psi. which includes plasticized polyvinyl chloride (PVC), low density polyethylene (LDPE), thermoplastic rubber, ethylene-ethyl acrylate, ethylene-butylene copolymer, polybutylene and copolymers thereof, ethylene-propylene copolymers, chlorinated polypropylene, chlorinated polybutylene or mixtures of those.

Many applications for such fibers require varying degrees of softness or stiffness and have different operating temperature requirements depending upon the application. For instance USP 5,068,141 (Kubo et al.) discloses making nonwoven fabrics from continuous heat bonded filaments of certain heterogeneously branched LLDPE having specified heats of fusion.

The present invention relates to fibers and fabricated articles therefrom prepared from polymer compositions which comprise at least one substantially random interpolymer comprising polymer units derived from one or more  $\alpha$ -olefin monomers with one or more vinyl or vinylidene aromatic monomers and/or a hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers or blends therefrom. Unique to these novel materials is the ability to precisely tune both the glass transition process (location, amplitude and width of transition) in the vicinity of the ambient temperature range, and the stiffness and modulus of the material in its final state. Both these factors can be controlled by varying the relative amount of  $\alpha$ -olefin(s) and vinyl or vinylidene aromatic and/or hindered aliphatic vinyl or vinylidene monomers in the final interpolymer or blend therefrom. Further variation in the Tg of the polymer composition used in the present invention can be introduced by variation of the type of component blended with the substantially random interpolymer including the presence of one or more tackifiers in the final formulation. This control of the Tg and modulus allows the stiffness or softness of the fiber to be varied to suit a given application.

We have discovered new fibers, fabrics and articles fabricated therefrom. These fibers and fabrics are made from novel substantially random interpolymers of  $\alpha$ -olefins and vinyl or vinylidene aromatic and/or hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers or blends therefrom. These interpolymers have a processability in fiber and fabric processes similar to homogeneous and heterogeneously branched

linear low density polyethylene, which means that the new fibers and fabrics can be produced on the conventional equipment used for the various synthetic fiber or fabric processes (for example, continuous wound filament, spun bond, and melt blown). The present invention pertains to fibers comprising;

- 5 (A) from about 50 to 100 wt percent (based on the combined weights of Components A and B) of at least one substantially random interpolymer having an  $I_2$  of from 0.1 to 1,000 g/10 min, a density greater than 0.9300 g/cm<sup>3</sup>, and an  $M_w/M_n$  of 1.5 to 20; which comprises;
- 10 (1) from 0.5 to 65 mol percent of polymer units derived from;
- (i) at least one vinyl or vinylidene aromatic monomer, or
- (ii) at least one hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or
- (iii) a combination of at least one aromatic vinyl or vinylidene monomer and at least one hindered aliphatic or
- 15 cycloaliphatic vinyl or vinylidene monomer, and
- (2) from 35 to 99.5 mol percent of polymer units derived from ethylene or at least one C<sub>3-20</sub>  $\alpha$ -olefin or a combination thereof; and
- (B) from 0 to 50 percent by weight (based on the combined weights of Components A and B) of at least one tackifier.

20 The fibers and fabrics and fabricated articles of the present invention show good elasticity, abrasion resistance, good viscoelastic properties such as resiliency, and possess both styrenic and olefinic functionality providing compatability with other styrenic-based materials and enabling their use as processing aids. For the fibers having a Tg close to body temperature, fabrics and clothing or other articles comprising

25 said fibers and for use on the human body show excellent body conformability.

Thus the fibers of the present invention have applications such as chemical separation membranes, dust masks, carpet fibers, elastic fibers, wigs, doll hair, personal/feminine hygiene applications, diapers, athletic sportswear, shin pads, wrinkle free and form-fitting apparel, upholstery, and medical applications including, but not

30 restricted to, surgical masks, bandages, gamma sterilizable fibers.

### Definitions

All references herein to elements or metals belonging to a certain Group refer to the Periodic Table of the Elements published and copyrighted by CRC Press, Inc., 1989. Also any reference to the Group or Groups shall be to the Group or Groups as reflected in this Periodic Table of the Elements using the IUPAC system for numbering groups.

Any numerical values recited herein include all values from the lower value to the upper value in increments of one unit provided that there is a separation of at least 2 units between any lower value and any higher value. As an example, if it is stated that the amount of a component or a value of a process variable such as, for example, temperature, pressure, time is, for example, from 1 to 90, preferably from 20 to 80, more preferably from 30 to 70, it is intended that values such as 15 to 85, 22 to 68, 43 to 51, 30 to 32 etc. are expressly enumerated in this specification. For values which are less than one, one unit is considered to be 0.0001, 0.001, 0.01 or 0.1 as appropriate. These are only examples of what is specifically intended and all possible combinations of numerical values between the lowest value and the highest value enumerated are to be considered to be expressly stated in this application in a similar manner.

The term "hydrocarbyl" as employed herein means any aliphatic, cycloaliphatic, aromatic, aryl substituted aliphatic, aryl substituted cycloaliphatic, aliphatic substituted aromatic, or aliphatic substituted cycloaliphatic groups.

The term "hydrocarbyloxy" means a hydrocarbyl group having an oxygen linkage between it and the carbon atom to which it is attached.

The term "interpolymer" is used herein to indicate a polymer wherein at least two different monomers are polymerized to make the interpolymer. This includes copolymers, terpolymers, etc.

The term "substantially random" (in the substantially random interpolymer comprising polymer units derived from one or more  $\alpha$ -olefin monomers with one or more vinyl or vinylidene aromatic monomers and/or one or more hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers) as used herein means that the distribution of the monomers of said interpolymer can be described by the Bernoulli statistical

model or by a first or second order Markovian statistical model, as described by J. C. Randall in POLYMER SEQUENCE DETERMINATION, Carbon-13 NMR Method, Academic Press New York, 1977, pp. 71-78. Preferably, substantially random interpolymers do not contain more than 15 percent of the total amount of vinyl or vinylidene aromatic monomer in blocks of vinyl or vinylidene aromatic monomer of more than 3 units. This means that in the carbon-13 NMR spectrum of the substantially random interpolymers the peak areas corresponding to the main chain methylene and methine carbons representing either meso diad sequences or racemic diad sequences should not exceed 75 percent of the total peak area of the main chain methylene and methine carbons.

#### The Fibers and Fabrics of the Present Invention

Fibers are often classified in terms of their diameter which can be measured and reported in a variety of fashions. Generally, fiber diameter is measured in denier per filament. Denier is a textile term which is defined as the grams of the fiber per 9000 meters of that fiber's length. Monofilament generally refers to an extruded strand having a denier per filament greater than 15, usually greater than 30. Fine denier fiber generally refers to fiber having a denier of about 15 or less. Microdenier (aka microfiber) generally refers to fibers having a diameter of less than about 1 denier. The fiber can also be classified by the process by which it is made, such as monofilament, continuous wound fine filament, staple or short cut fiber, spun bond, and melt blown fiber. Fiber can also be classified by the number of regions or domains in the fiber.

The fibers of the present invention include the various homofil fibers made from the substantially random interpolymers or blend compositions therefrom. Homofil fibers are those fibers which have a single region (domain) and do not have other distinct polymer regions (as do bicomponent fibers). These homofil fibers include staple fibers, spunbond fibers or melt blown fibers (using, for example, systems as disclosed in USP 4,340,563 (Appel et al.), USP 4,663,220 (Wisneski et al.), USP 4,668,566 (Braun), or USP 4,322,027 (Reba), and gel spun fibers (for example, the system disclosed in USP 4,413,110 (Kavesh et al.). Staple fibers can be melt spun

(that is, they can be extruded into the final fiber diameter directly without additional drawing), or they can be melt spun into a higher diameter and subsequently hot or cold drawn to the desired diameter using conventional fiber drawing techniques. The novel staple fibers disclosed herein can also be used as bonding fibers, especially where the novel fibers have a lower melting point than the surrounding matrix fibers. In a bonding fiber application, the bonding fiber is typically blended with other matrix fibers and the entire structure is subjected to heat, where the bonding fiber melts and bonds the surrounding matrix fiber. Typical matrix fibers which benefit from use of the novel fibers of the present invention includes, but is not limited to, synthetic fibers, made from fiber glass, poly(ethylene terephthalate), polypropylene, nylon, heterogeneously branched polyethylene, linear and substantially linear ethylene interpolymers or polyethylene homopolymers. The matrix fibers can also comprise natural fibers such as silk, wool, and cotton. The diameter of the matrix fiber can vary depending upon the end use application.

The fibers of the present invention also include the various composite fibers which can comprise the novel substantially random interpolymers and a second polymer component. This second polymer component can be an ethylene or  $\alpha$ -olefin homopolymer or interpolymer; an ethylene/propylene rubber (EPM), ethylene/propylene diene monomer terpolymer (EPDM), isotactic polypropylene; a styrene/ethylene-butene copolymer, a styrene/ethylene-propylene copolymer, a styrene/ethylene-butene/styrene (SEBS) copolymer, a styrene/ethylene-propylene/styrene (SEPS) copolymer; the acrylonitrile-butadiene-styrene (ABS) polymers, styrene-acrylonitrile (SAN), high impact polystyrene, polyisoprene, polybutadiene, natural rubbers, ethylene/propylene rubbers, ethylene/propylene diene (EPDM) rubbers, styrene/butadiene rubbers, thermoplastic polyurethanes, epoxies, vinyl ester resins, polyurethanes, phenolic resins, homopolymers or copolymers of vinyl chloride or vinylidene chloride, poly(methylmethacrylate), polyester, nylon-6, nylon-6,6, poly(acetal); poly(amide), poly(arylate), poly(carbonate), poly(butylene) and polybutylene, polyethylene terephthalates; or blend compositions therefrom. Preferably the second polymer component is an



ethylene or  $\alpha$ -olefin homopolymer or interpolymers, wherein said  $\alpha$ -olefin has from 3 to 20 carbon atoms, and polyethylene terephthalates.

The most prevalent composite fibers are the bicomponent fibers which have two polymers in a co-continuous phase. Examples of such bicomponent fiber configurations and shapes include sheath/core fibers in which the perimeter shape is round, oval, delta, trilobal, triangular, dog-boned, or flat or hollow configurations. Other types of bicomponent fibers within the scope of the invention include such structures as segmented pies, as well as side-by-side fibers (for example, fibers having separate regions of polymers, wherein the substantially random interpolymers comprises at least a portion of the fiber's surface). Also included are the "islands in the sea" bicomponent fibers in which a cross section of the fiber has a main matrix of the first polymer component dispersed across which are extruded domains of a second polymer. On viewing a cross section of such a fiber, the main polymer matrix appears as a "sea" in which the domains of the second polymer component appear as islands.

The bicomponent fibers of the present invention can be prepared by coextruding a substantially random interpolymers in at least one portion of the fiber and a second polymer component in at least one other portion of the fiber. For all configurations of a bicomponent fiber in which the sheath concentrically surrounds the core, the substantially random interpolymers can be in either the sheath or the core. Different substantially random interpolymers can also be used independently as the sheath and the core in the same fiber, and especially where the sheath component has a lower melting point than the core component. In the case of segmented pie configurations, one or more of the segments can comprise the substantially random interpolymers. In the case of an "island in the sea" configuration, either the islands or the matrix can comprise the substantially random interpolymers. The bicomponent fiber can be formed under melt blown, spunbond, continuous filament or staple fiber manufacturing conditions.

Finishing operations can optionally be performed on the fibers of the present invention. For example, the fibers can be texturized by mechanically crimping or

forming such as described in Textile Fibers, Dyes, Finishes, and Processes: A Concise Guide, by Howard L. Needles, Noyes Publications, 1986, pp. 17-20.

The polymer compositions used to make the fibers of the present invention or the fibers themselves may be modified by various cross-linking processes using curing methods at any stage of the fiber preparation including, but not limited to, before during, and after drawing at either elevated or ambient temperatures. Such cross-linking processes include, but are not limited to, peroxide-, silane-, sulfur-, radiation-, or azide-based cure systems. A full description of the various cross-linking technologies is described in copending U.S. Patent Application No's 08/921,641 and 08/921,642 both filed on August 27, 1997.

Dual cure systems, which use a combination of heat, moisture cure, and radiation steps, may be effectively employed. Dual cure systems are disclosed and claimed in U. S. Patent Application Serial No. 536,022, filed on September 29, 1995, in the names of K. L. Walton and S. V. Karande. For instance, it may be desirable to employ peroxide crosslinking agents in conjunction with silane crosslinking agents, peroxide crosslinking agents in conjunction with radiation, sulfur-containing crosslinking agents in conjunction with silane crosslinking agents, etc.

The polymer compositions may also be modified by various cross-linking processes including, but not limited to the incorporation of a diene component as a termonomer in its preparation and subsequent cross linking by the aforementioned methods and further methods including vulcanization via the vinyl group using sulfur for example as the cross linking agent.

The fibers of the present invention may be surface functionalized by methods including, but not limited to sulfonation, chlorination using chemical treatments for permanent surfaces or incorporating a temporary coating using the various well known spin finishing processes.

Fabrics made from such novel fibers include both woven and nonwoven fabrics. Nonwoven fabrics can be made variously, including spunlaced (or hydrodynamically entangled) fabrics as disclosed in USP 3,485,706 (Evans) and USP 4,939,016 (Radwanski et al.); by carding and thermally bonding homofil or bicomponent staple

fibers by spunbonding homofil or bicomponent fibers in one continuous operation; or by melt blowing homofil or bicomponent fibers into fabric and subsequently calendering or thermally bonding the resultant web. Other structures made from such fibers are also included within the scope of the invention, including for example, blends  
5 of these novel fibers with other fibers (for example, poly(ethylene terephthalate) (PET) or cotton or wool or polyester).

Woven fabrics can also be made which comprise the fibers of the present invention. The various woven fabric manufacturing techniques are well known to those skilled in the art and the disclosure is not limited to any particular method. Woven  
10 fabrics are typically stronger and more heat resistant and are thus used typically in durable, non-disposable applications as for example in the woven blends with polyester and polyester cotton blends. The woven fabrics comprising the fibers of the present invention can be used in applications including but not limited to, upholstery, athletic apparel, carpet, fabrics, bandages.

15 The novel fibers and fabrics disclosed herein can also be used in various structures as described in USP 2,957,512 (Wade). Attachment of the novel fibers and/or fabric to fibers, fabrics or other structures can be done with melt bonding or with adhesives. Gathered or shirred structures can be produced from the new fibers and/or fabrics and other components by pleating the other component (as described in USP  
20 '512) prior to attachment, prestretching the novel fiber component prior to attachment, or heat shrinking the novel fiber component after attachment.

The novel fibers described herein also can be used in a spunlaced (or hydrodynamically entangled) process to make novel structures. For example, USP  
25 4,801,482 (Goggans), discloses a sheet which can now be made with the novel fibers/fabric described herein.

Composites that utilize very high molecular weight linear polyethylene or copolymer polyethylene also benefit from the novel fibers disclosed herein. For example, for the novel fibers that have a low melting point, such that in a blend of the novel fibers and very high molecular weight polyethylene fibers (for example,  
30 Spectra<sup>TM</sup> fibers made by Allied Chemical) as described in USP 4,584,347 (Harpell et

al.), the lower melting fibers bond the high molecular weight polyethylene fibers without melting the high molecular weight fibers, thus preserving the high strength and integrity of the high molecular weight fiber.

The fibers and fabrics can have additional materials which do not materially affect their properties. Such useful nonlimiting additive materials include pigments, antioxidants, stabilizers, surfactants (for example, as disclosed in USP 4,486,552 (Niemann), USP 4,578,414 (Sawyer et al.) or USP 4,835,194 (Bright et al.).

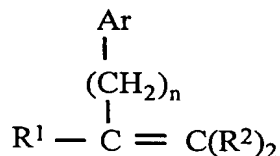
#### The Substantially Random Interpolymers

The interpolymers used to prepare the fibers of the present invention include interpolymers prepared by polymerizing one or more  $\alpha$ -olefins with one or more vinyl or vinylidene aromatic monomers and/or one or more hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers, and optionally other polymerizable monomers.

Suitable  $\alpha$ -olefins include for example,  $\alpha$ -olefins containing from 2 to 20, preferably from 2 to 12, more preferably from 2 to 8 carbon atoms. Particularly suitable are ethylene, propylene, butene-1, 4-methyl-1-pentene, hexene-1 or octene-1 or ethylene in combination with one or more of propylene, butene-1, 4-methyl-1-pentene, hexene-1 or octene-1. These  $\alpha$ -olefins do not contain an aromatic moiety.

Other optional polymerizable ethylenically unsaturated monomer(s) include strained ring olefins such as norbornene and  $C_{1-10}$  alkyl or  $C_{6-10}$  aryl substituted norbornenes, with an exemplary interpolymer being ethylene/styrene/norbornene.

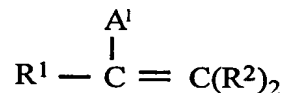
Suitable vinyl or vinylidene aromatic monomers which can be employed to prepare the interpolymers include, for example, those represented by the following formula:



wherein  $\text{R}^1$  is selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; each  $\text{R}^2$  is

independently selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; Ar is a phenyl group or a phenyl group substituted with from 1 to 5 substituents selected from the group consisting of halo, C<sub>1-4</sub>-alkyl, and C<sub>1-4</sub>-haloalkyl; and n has a value from zero to 4, preferably from zero to 2, most preferably zero. Exemplary vinyl aromatic monomers include styrene, vinyl toluene,  $\alpha$ -methylstyrene, t-butyl styrene, chlorostyrene, including all isomers of these compounds,. Particularly suitable such monomers include styrene and lower alkyl- or halogen-substituted derivatives thereof. Preferred monomers include styrene,  $\alpha$ -methyl styrene, the lower alkyl- (C<sub>1</sub> - C<sub>4</sub>) or phenyl-ring substituted derivatives of styrene, such as for example, ortho-, meta-, and para-methylstyrene, the ring halogenated styrenes, para-vinyl toluene or mixtures thereof,. A more preferred aromatic vinylmonomer is styrene.

By the term "hindered aliphatic or cycloaliphatic vinyl or vinylidene compounds", it is meant addition polymerizable vinyl or vinylidene monomers corresponding to the formula:



wherein A<sup>1</sup> is a sterically bulky, aliphatic or cycloaliphatic substituent of up to 20 carbons, R<sup>1</sup> is selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; each R<sup>2</sup> is independently selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; or alternatively R<sup>1</sup> and A<sup>1</sup> together form a ring system. By the term "sterically bulky" is meant that the monomer bearing this substituent is normally incapable of addition polymerization by standard Ziegler-Natta polymerization catalysts at a rate comparable with ethylene polymerizations. Preferred hindered aliphatic or cycloaliphatic vinyl or vinylidene compounds are monomers in which one of the carbon atoms bearing ethylenic unsaturation is tertiary or quaternary substituted. Examples of such substituents include cyclic aliphatic groups such as cyclohexyl, cyclohexenyl, cyclooctenyl, or ring alkyl or aryl substituted derivatives thereof, tert-butyl, and

norbornyl. Most preferred hindered aliphatic or cycloaliphatic vinyl or vinylidene compounds are the various isomeric vinyl- ring substituted derivatives of cyclohexene and substituted cyclohexenes, and 5-ethylidene-2-norbornene. Especially suitable are 1-, 3-, and 4-vinylcyclohexene.

5       The substantially random interpolymers may be modified by typical grafting, hydrogenation, functionalizing, or other reactions well known to those skilled in the art. The polymers may be readily sulfonated or chlorinated to provide functionalized derivatives according to established techniques.

10       The substantially random interpolymers may also be modified by various cross-linking processes including, but not limited to peroxide-, silane-, sulfur-, radiation-, or azide-based cure systems. A full description of the various cross-linking technologies is described in copending U.S. Patent Application No's 08/921,641 and 08/921,642 both filed on August 27, 1997.

15       Dual cure systems, which use a combination of heat, moisture cure, and radiation steps, may be effectively employed. Dual cure systems are disclosed and claimed in U. S. Patent Application Serial No. 536,022, filed on September 29, 1995, in the names of K. L. Walton and S. V. Karande. For instance, it may be desirable to employ peroxide crosslinking agents in conjunction with silane crosslinking agents, peroxide crosslinking agents in conjunction with radiation, sulfur-containing  
20       crosslinking agents in conjunction with silane crosslinking agents, etc.

25       The substantially random interpolymers may also be modified by various cross-linking processes including, but not limited to, the incorporation of a diene component as a termonomer in its preparation and subsequent cross linking by the aforementioned methods and further methods including vulcanization via the vinyl group using sulfur for example as the cross linking agent.

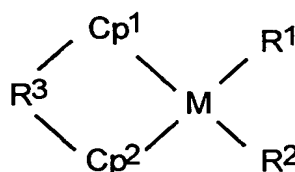
      One method of preparation of the substantially random interpolymers includes polymerizing a mixture of polymerizable monomers in the presence of one or more metallocene or constrained geometry catalysts in combination with various cocatalysts.

30       The substantially random interpolymers can be prepared as described in EP-A-0,416,815 and US Patent No. 5,703,187 by Francis Timmers. Preferred operating

conditions for such polymerization reactions are pressures from atmospheric up to 3000 atmospheres and temperatures from -30°C to 200°C. Polymerizations and unreacted monomer removal at temperatures above the autopolymerization temperature of the respective monomers may result in formation of some amounts of homopolymer  
5 polymerization products resulting from free radical polymerization.

Examples of suitable catalysts and methods for preparing the substantially random interpolymers are disclosed in U.S. Application Serial No. 545,403, filed July 3, 1990 (EP-A-416,815); U.S. Application Serial No. 702,475, filed May 20, 1991 (EP-A-514,828); U.S. Application Serial No. 876,268, filed May 1, 1992, (EP-A-520,732);  
10 U.S. Application Serial No. 241,523, filed May 12, 1994; as well as U.S. Patents: 5,055,438; 5,057,475; 5,096,867; 5,064,802; 5,132,380; 5,189,192; 5,321,106; 5,347,024; 5,350,723; 5,374,696; and 5,399,635.

The substantially random  $\alpha$ -olefin/vinyl or vinylidene aromatic interpolymers can also be prepared by the methods described in JP 07/278230 employing compounds  
15 shown by the general formula



where  $Cp^1$  and  $Cp^2$  are cyclopentadienyl groups, indenyl groups, fluorenyl groups, or substituents of these, independently of each other;  $R^1$  and  $R^2$  are hydrogen atoms, halogen atoms, hydrocarbon groups with carbon numbers of 1-12, alkoxyl groups, or  
20 aryloxyl groups, independently of each other; M is a group IV metal, preferably Zr or Hf, most preferably Zr; and  $R^3$  is an alkylene group or silanediyl group used to cross-link  $Cp^1$  and  $Cp^2$ ).

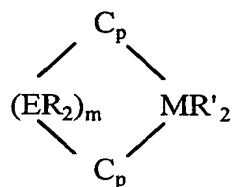
The substantially random  $\alpha$ -olefin/vinyl or vinylidene aromatic interpolymers can also be prepared by the methods described by John G. Bradfute et al. (W. R. Grace & Co.) in WO 95/32095; by R. B. Pannell (Exxon Chemical Patents, Inc.) in WO  
25 94/00500; and in Plastics Technology, p. 25 (September 1992).

Also suitable are the substantially random interpolymers which comprise at least one  $\alpha$ -olefin/vinyl aromatic/vinyl aromatic/ $\alpha$ -olefin tetrad disclosed in U. S.

Application No. 08/708,809 filed September 4, 1996 by Francis J. Timmers et al.

These interpolymers contain additional signals in their carbon-13 NMR spectra with intensities greater than three times the peak to peak noise. These signals appear in the chemical shift range 43.70 - 44.25 ppm and 38.0 - 38.5 ppm. Specifically, major peaks  
 5 are observed at 44.1, 43.9, and 38.2 ppm. A proton test NMR experiment indicates that the signals in the chemical shift region 43.70 - 44.25 ppm are methine carbons and the signals in the region 38.0 - 38.5 ppm are methylene carbons.

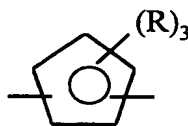
It is believed that these new signals are due to sequences involving two head-to-tail vinyl aromatic monomer insertions preceded and followed by at least one  $\alpha$ -olefin  
 10 insertion, for example an ethylene/styrene/styrene/ethylene tetrad wherein the styrene monomer insertions of said tetrads occur exclusively in a 1,2 (head to tail) manner. It is understood by one skilled in the art that for such tetrads involving a vinyl aromatic monomer other than styrene and an  $\alpha$ -olefin other than ethylene that the ethylene/vinyl aromatic monomer/vinyl aromatic monomer/ethylene tetrad will give rise to similar  
 15 carbon-13 NMR peaks but with slightly different chemical shifts. These interpolymers can be prepared by conducting the polymerization at temperatures of from about -30°C to 250°C in the presence of such catalysts as those represented by the formula



wherein: each  $C_p$  is independently, each occurrence, a substituted cyclopentadienyl  
 20 group  $\pi$ -bound to M; E is C or Si; M is a group IV metal, preferably Zr or Hf, most preferably Zr; each R is independently, each occurrence, H, hydrocarbyl, silahydrocarbyl, or hydrocarbylsilyl, containing up to 30 preferably from 1 to 20 more preferably from 1 to 10 carbon or silicon atoms; each R' is independently, each  
 occurrence, H, halo, hydrocarbyl, hydrocarbyloxy, silahydrocarbyl, hydrocarbylsilyl  
 25 containing up to 30 preferably from 1 to 20 more preferably from 1 to 10 carbon or silicon atoms or two R' groups together can be a  $C_{1-10}$  hydrocarbyl substituted 1,3-butadiene; m is 1 or 2; and optionally, but preferably in the presence of an activating



cocatalyst. Particularly suitable substituted cyclopentadienyl groups include those illustrated by the formula:



wherein each R is independently, each occurrence, H, hydrocarbyl, silahydrocarbyl, or hydrocarbylsilyl, containing up to 30 preferably from 1 to 20 more preferably from 1 to 10 carbon or silicon atoms or two R groups together form a divalent derivative of such group. Preferably, R independently each occurrence is (including where appropriate all isomers) hydrogen, methyl, ethyl, propyl, butyl, pentyl, hexyl, benzyl, phenyl or silyl or (where appropriate) two such R groups are linked together forming a fused ring system such as indenyl, fluorenyl, tetrahydroindenyl, tetrahydrofluorenyl, or octahydrofluorenyl.

Particularly preferred catalysts include, for example, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl))zirconium dichloride, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl))zirconium 1,4-diphenyl-1,3-butadiene, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl))zirconium di-C<sub>1-4</sub> alkyl, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl))zirconium di-C<sub>1-4</sub> alkoxide, or any combination thereof.

It is also possible to use the titanium-based constrained geometry catalysts, [N-(1,1-dimethylethyl)-1,1-dimethyl-1-[(1,2,3,4,5-η)-1,5,6,7-tetrahydro-s-indacen-1-yl]silanaminato(2-)-N]titanium dimethyl; (1-indenyl)(tert-butylamido)dimethylsilane titanium dimethyl; ((3-tert-butyl)(1,2,3,4,5-η)-1-indenyl)(tert-butylamido)dimethylsilane titanium dimethyl; and ((3-iso-propyl)(1,2,3,4,5-η)-1-indenyl)(tert-butyl amido)dimethylsilane titanium dimethyl, or any combination thereof.

Further preparative methods for the interpolymers used in the present invention have been described in the literature. Longo and Grassi (Makromol. Chem., Volume 191, pages 2387 to 2396 [1990]) and D'Anniello et al. (Journal of Applied Polymer Science, Volume 58, pages 1701-1706 [1995]) reported the use of a catalytic system based on methylalumoxane (MAO) and cyclopentadienyltitanium trichloride (CpTiCl<sub>3</sub>) to prepare an ethylene-styrene copolymer. Xu and Lin (Polymer Preprints, Am. Chem.

Soc., Div. Polym. Chem.) Volume 35, pages 686,687 [1994]) have reported copolymerization using a  $\text{MgCl}_2/\text{TiCl}_4/\text{NdCl}_3/\text{Al}(\text{iBu})_3$  catalyst to give random copolymers of styrene and propylene. Lu et al (Journal of Applied Polymer Science, Volume 53, pages 1453 to 1460 [1994]) have described the copolymerization of ethylene and styrene using a  $\text{TiCl}_4/\text{NdCl}_3/\text{MgCl}_2/\text{Al}(\text{Et})_3$  catalyst. Sernetz and Mulhaupt, (Macromol. Chem. Phys., v. 197, pp 1071-1083, 1997) have described the influence of polymerization conditions on the copolymerization of styrene with ethylene using  $\text{Me}_2\text{Si}(\text{Me}_4\text{Cp})(\text{N-tert-butyl})\text{TiCl}_2/\text{methylaluminumoxane}$  Ziegler-Natta catalysts. Copolymers of ethylene and styrene produced by bridged metallocene catalysts have been described by Arai, Toshiaki and Suzuki (Polymer Preprints, Am. Chem. Soc., Div. Polym. Chem.) Volume 38, pages 349, 350 [1997]). The manufacture of  $\alpha$ -olefin/vinyl aromatic monomer interpolymers such as propylene/styrene and butene/styrene are described in United States patent number 5,244,996, issued to Mitsui Petrochemical Industries Ltd or United States patent number 5,652,315 also issued to Mitsui Petrochemical Industries Ltd or as disclosed in DE 197 11 339 A1 to Denki Kagaku Kogyo KK.

While preparing the substantially random interpolpolymer, an amount of atactic vinyl or vinylidene aromatic homopolymer may be formed due to homopolymerization of the vinyl or vinylidene aromatic monomer at elevated temperatures. The presence of vinyl or vinylidene aromatic homopolymer is in general not detrimental for the purposes of the present invention and can be tolerated. The vinyl or vinylidene aromatic homopolymer may be separated from the interpolpolymer, if desired, by extraction techniques such as selective precipitation from solution with a non solvent for either the interpolpolymer or the vinyl or vinylidene aromatic homopolymer. For the purpose of the present invention it is preferred that no more than 20 weight percent, preferably less than 15 weight percent based on the total weight of the interpolpolymers of atactic vinyl or vinylidene aromatic homopolymer is present.

#### Blend Compositions Comprising the Substantially Random Interpolymers

The present invention also provides fibers prepared from blends of the substantially random  $\alpha$ -olefin/ vinyl or vinylidene interpolymers with one or more

other polymer components which span a wide range of compositions. When the fiber is prepared using a blend composition comprising another polymer component, it is understood that said fiber can be prepared directly from the blended polymer composition or be prepared by combining one or more pre-formed fibers of the substantially random interpolymer and the other polymer component. When the fiber has a bicomponent structure, then either the core or the sheath can comprise either the substantially random interpolymer and the other polymer component.

The other polymer component of the blend can include, but is not limited to, one or more of an engineering thermoplastic, an  $\alpha$ -olefin homopolymer or interpolymer, a thermoplastic olefin, a styrenic block copolymer, a styrenic copolymer, an elastomer, a thermoset polymer, or a vinyl halide polymer.

#### The Engineering Thermoplastic

The third edition of the Kirk-Othmer Encyclopedia of Science and Technology defines engineering plastics as thermoplastic resins, neat or unreinforced or filled, which maintain dimensional stability and most mechanical properties above 100°C and below 0°C. The terms "engineering plastics" and "engineering thermoplastics", can be used interchangeably. Engineering Thermoplastics include acetal and acrylic resins such as polymethylmethacrylate (PMMA), polyamides (for example nylon-6, nylon 6,6), polyimides, polyetherimides, cellulose, polyesters, poly(arylate), aromatic polyesters, poly(carbonate), poly(butylene) and polybutylene and polyethylene terephthalates, liquid crystal polymers, and selected polyolefins, blends, or alloys of the foregoing resins, and some examples from other resin types (including for example polyethers) high temperature polyolefins such as polycyclopentanes, its copolymers, and polymethylpentane.).

Most acrylic resins derive from the peroxide-catalyzed free radical polymerization of methyl methacrylate (MMA) to make polymethylmethacrylate (PMMA). As described by H. Luke in Modern Plastics Encyclopedia, 1989, pps 20-21, MMA is usually copolymerized with other acrylates such as methyl- or ethyl acrylate using four basic polymerization processes, bulk, suspension, emulsion and solution.

Acrylics can also be modified with various ingredients including styrene, butadiene, vinyl and alkyl acrylates. Acrylics known as PMMA have ASTM grades and specifications. Grades 5, 6 and 8 vary mainly in deflection temperature under load (DTL) requirements. Grade 8 requires a tensile strength of 9,000 psi vs 8,000 psi for  
5 Grades 5 and 6. The DTL varies from a minimum requirement of 153°F to a maximum of 189°F, under a load of 264 p.s.i. Certain grades have a DTL of 212°F. Impact-modified grades range from an Izod impact of 1.1 to 2.0 ft .lb/in for non-weatherable transparent materials. The opaque impact-modified grades can have Izod impact values as high as 5.0 ft lb/in.

10 We have surprisingly found that when PMMA is incorporated into the polymer compositions used to prepare the fibers of the present invention a number of unexpected advantages are observed. Thus when the structure or fabricated article comprises a fiber, the addition of up to 20, preferably up to 10 wt percent of acrylic resin in the polymer composition used to prepare said fiber can result in an increase of  
15 the gloss of the fiber and an improvement in the fiber handling characteristics ( that is the fibers have a lower tendency to stick together which greatly facilitates such procedures as fiber carding and/or combing).

Also preferred as the other polymer component of the blends used to prepare the fibers of the present invention are the polyesters.

20 Polyesters may be made by the self-esterification of hydroxycarboxylic acids, or by direct esterification, which involves the step-growth reaction of a diol with a dicarboxylic acid with the resulting elimination of water, giving a polyester with an -[AABB]- repeating unit. The reaction may be run in bulk or in solution using an inert high boiling solvent such as xylene or chlorobenzene with azeotropic removal of water.

25 Alternatively, but in like manner, ester-forming derivatives of a dicarboxylic acid can be heated with a diol to obtain polyesters in an ester interchange reaction. Suitable acid derivatives for such purpose are alkyl esters, halides, salts or anhydrides of the acid. Preparation of polyarylates, from a bisphenol and an aromatic diacid, can be conducted in an interfacial system which is essentially the same as that used for the  
30 preparation of polycarbonate.

Polyesters can also be produced by a ring-opening reaction of cyclic esters or C<sub>4</sub>-C<sub>7</sub> lactones, for which organic tertiary amine bases phosphines and alkali and alkaline earth metals, hydrides and alkoxides can be used as initiators.

Suitable reactants for making the polyester used in this invention, in addition to  
5 hydroxycarboxylic acids, are diols and dicarboxylic acids either or both of which can be aliphatic or aromatic. A polyester which is a poly(alkylene alkanedicarboxylate), a poly(alkylene arylenedicarboxylate), a poly(arylene alkanedicarboxylate), or a poly(arylene arylenedicarboxylate) is therefore appropriate for use herein. Alkyl  
10 portions of the polymer chain can be substituted with, for example, halogens, C<sub>1</sub>-C<sub>8</sub> alkoxy groups or C<sub>1</sub>-C<sub>8</sub> alkyl side chains and can contain divalent heteroatomic groups (such as -O-, -Si-, -S- or -SO<sub>2</sub>-) in the paraffinic segment of the chain. The chain can also contain unsaturation and C<sub>6</sub>-C<sub>10</sub> non-aromatic rings. Aromatic rings can contain substituents such as halogens, C<sub>1</sub>-C<sub>8</sub> alkoxy or C<sub>1</sub>-C<sub>8</sub> alkyl groups, and can be joined to the polymer backbone in any ring position and directly to the alcohol or acid  
15 functionality or to intervening atoms.

Typical aliphatic diols used in ester formation are the C<sub>2</sub>-C<sub>10</sub> primary and secondary glycols, such as ethylene-, propylene-, and butylene glycol. Alkanedicarboxylic acids frequently used are oxalic acid, adipic acid and sebacic acid. Diols which contain rings can be, for example, a 1,4-cyclohexylenyl glycol or a 1,4-  
20 cyclohexane-dimethylene glycol, resorcinol, hydroquinone, 4,4'-thiodiphenol, bis-(4-hydroxyphenyl)sulfone, a dihydroxynaphthalene, a xylylene diol, or can be one of the many bisphenols such as 2,2-bis-(4-hydroxyphenyl)propane. Aromatic diacids include, for example, terephthalic acid, isophthalic acid, naphthalenedicarboxylic acid, diphenyletherdicarboxylic acid, diphenyldicarboxylic acid, diphenylsulfonedicarboxylic  
25 acid, diphenoxyethanedicarboxylic acid.

In addition to polyesters formed from one diol and one diacid only, the term "polyester" as used herein includes random, patterned or block copolyesters, for example those formed from two or more different diols and/or two or more different diacids, and/or from other divalent heteroatomic groups. Mixtures of such  
30 copolyesters, mixtures of polyesters derived from one diol and diacid only, and

mixtures of members from both of such groups, are also all suitable for use in this invention, and are all included in the term "polyester". For example, use of cyclohexanedimethanol together with ethylene glycol in esterification with terephthalic acid forms a clear, amorphous copolyester of particular interest. Also contemplated are  
5 liquid crystalline polyesters derived from mixtures of 4-hydroxybenzoic acid and 2-hydroxy-6-naphthoic acid; or mixtures of terephthalic acid, 4-hydroxybenzoic acid and ethylene glycol; or mixtures of terephthalic acid, 4-hydroxybenzoic acid and 4,4'-dihydroxybiphenyl.

Aromatic polyesters, those prepared from an aromatic diacid, such as the  
10 poly(alkylene arylenedicarboxylates) polyethylene terephthalate and polybutylene terephthalate, or mixtures thereof, are particularly useful in this invention. A polyester suitable for use herein may have an intrinsic viscosity of 0.4 to 1.2, although values outside this range are permitted as well.

Methods and materials useful for the production of polyesters, as described  
15 above, are discussed in greater detail in Whinfield, USP 2,465,319, Pengilly, USP 3,047,539, Schwarz, USP 3,374,402, Russell, USP 3,756,986 and East, USP 4,393,191.

#### The $\alpha$ -Olefin Homopolymers and Interpolymers

The  $\alpha$ -olefin homopolymers and interpolymers comprise polypropylene,  
20 propylene/C<sub>4</sub>-C<sub>20</sub>  $\alpha$ -olefin copolymers, polyethylene, and ethylene/C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin copolymers, the interpolymers can be either heterogeneous ethylene/ $\alpha$ -olefin interpolymers or homogeneous ethylene/ $\alpha$ -olefin interpolymers, including the substantially linear ethylene/ $\alpha$ -olefin interpolymers.

Heterogeneous interpolymers are differentiated from the homogeneous  
25 interpolymers in that in the latter, substantially all of the interpolymer molecules have the same ethylene/comonomer ratio within that interpolymer, whereas heterogeneous interpolymers are those in which the interpolymer molecules do not have the same ethylene/comonomer ratio. The term "broad composition distribution" used herein describes the comonomer distribution for heterogeneous interpolymers and means that  
30 the heterogeneous interpolymers have a "linear" fraction and that the heterogeneous

interpolymers have multiple melting peaks (that is, exhibit at least two distinct melting peaks) by DSC. The heterogeneous interpolymers have a degree of branching less than or equal to 2 methyls/1000 carbons in 10 percent (by weight) or more, preferably more than 15 percent (by weight), and especially more than 20 percent (by weight). The  
5 heterogeneous interpolymers also have a degree of branching equal to or greater than 25 methyls/1000 carbons in 25 percent or less (by weight), preferably less than 15 percent (by weight), and especially less than 10 percent (by weight).

The Ziegler catalysts suitable for the preparation of the heterogeneous component of the current invention are typical supported, Ziegler-type catalysts which  
10 are particularly useful at the high polymerization temperatures of the solution process. Examples of such compositions are those derived from organomagnesium compounds, alkyl halides or aluminum halides or hydrogen chloride, and a transition metal compound. Examples of such catalysts are described in U.S. Pat Nos. 4,314,912 (Lowery, Jr. et al.), 4,547,475 (Glass et al.), and 4,612,300 (Coleman, III).

15 Suitable catalyst materials may also be derived from a inert oxide supports and transition metal compounds. Examples of such compositions suitable for use in the solution polymerization process are described in U.S. Pat No. 5,420,090 (Spencer. et al.).

The heterogeneous polymer component can be an  $\alpha$ -olefin homopolymer  
20 preferably polyethylene or polypropylene, or, preferably, an interpolymer of ethylene with at least one C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin and/or C<sub>4</sub>-C<sub>18</sub> diolefins. Heterogeneous copolymers of ethylene and 1-octene are especially preferred.

The relatively recent introduction of metallocene-based catalysts for ethylene/ $\alpha$ -olefin polymerization has resulted in the production of new ethylene interpolymers and  
25 new requirements for compositions containing these materials. Such polymers are known as homogeneous interpolymers and are characterized by their narrower molecular weight and composition distributions (defined as the weight percent of the polymer molecules having a comonomer content within 50 percent of the median total molar comonomer content) relative to, for example, traditional Ziegler catalyzed  
30 heterogeneous polyolefin polymers. Generally blown and cast film made with such

polymers are tougher and have better optical properties and heat sealability than film made with Ziegler Natta catalyzed LLDPE. It is known that metallocene LLDPE offers significant advantages over Ziegler Natta produced LLDPE's in cast film for pallet wrap applications, particularly improved on-pallet puncture resistance. Such  
5 metallocene LLDPE's however have a significantly poorer processability on the extruder than Ziegler Natta products.

The substantially linear ethylene/ $\alpha$ -olefin polymers and interpolymers of the present invention are herein defined as in U.S. Patent No. 5,272,236 and U.S. Patent No. 5,278,272 (Lai et al.). The substantially linear ethylene/ $\alpha$ -olefin polymers are also  
10 metallocene based homogeneous polymers, as the comonomer is randomly distributed within a given interpolymer molecule and wherein substantially all of the interpolymer molecules have the same ethylene/comonomer ratio within that interpolymer. Such polymers are unique however due to their excellent processability and unique rheological properties and high melt elasticity and resistance to melt fracture. These  
15 polymers can be successfully prepared in a continuous polymerization process using the constrained geometry metallocene catalyst systems.

The substantially linear ethylene/ $\alpha$ -olefin polymers and are those in which the comonomer is randomly distributed within a given interpolymer molecule and wherein substantially all of the interpolymer molecules have the same ethylene/comonomer  
20 ratio within that interpolymer.

The term "substantially linear" ethylene/ $\alpha$ -olefin interpolymer means that the polymer backbone is substituted with 0.01 long chain branches/1000 carbons to 3 long chain branches/1000 carbons, more preferably from 0.01 long chain branches/1000 carbons to 1 long chain branches/1000 carbons, and especially from 0.05 long chain  
25 branches/1000 carbons to 1 long chain branches/1000 carbons.

Long chain branching is defined herein as a chain length of at least one carbon more than two carbons less than the total number of carbons in the comonomer, for example, the long chain branch of an ethylene/octene substantially linear ethylene interpolymer is at least seven (7) carbons in length (that is, 8 carbons less 2 equals 6  
30 carbons plus one equals seven carbons long chain branch length). The long chain



branch can be as long as the same length as the length of the polymer back-bone. Long chain branching is determined by using  $^{13}\text{C}$  nuclear magnetic resonance (NMR) spectroscopy and is quantified using the method of Randall (Rev. Macromol. Chem. Phys., C29 (2&3), p. 285-297). Long chain branching, of course, is to be distinguished from short chain branches which result solely from incorporation of the comonomer, so for example the short chain branch of an ethylene/octene substantially linear polymer is six carbons in length, while the long chain branch for that same polymer is at least seven carbons in length.

The "rheological processing index" (PI) is the apparent viscosity (in kpoise) of a polymer measured by a gas extrusion rheometer (GER). The gas extrusion rheometer is described by M. Shida, R.N. Shroff and L.V. Cancio in Polymer Engineering Science, Vol. 17, no. 11, p. 770 (1977), and in "Rheometers for Molten Plastics" by John Dealy, published by Van Nostrand Reinhold Co. (1982) on page 97-99. All GER experiments are performed at a temperature of  $190^{\circ}\text{C}$ , at nitrogen pressures between 5250 to 500 psig using a 0.0296 inch diameter, 20:1 L/D die with an entrance angle of  $180^{\circ}$ . For the substantially linear ethylene/ $\alpha$ -olefin polymers described herein, the PI is the apparent viscosity (in kpoise) of a material measured by GER at an apparent shear stress of  $2.15 \times 10^6$  dyne/cm<sup>2</sup>. The novel substantially linear ethylene/ $\alpha$ -olefin interpolymers described herein preferably have a PI in the range of 0.01 kpoise to 50 kpoise, preferably 15 kpoise or less. The novel substantially linear ethylene/ $\alpha$ -olefin polymers described herein have a PI less than or equal to about 70 percent of the PI of a comparative linear ethylene/ $\alpha$ -olefin polymer at about the same  $I_2$  and  $M_w/M_n$ .

An apparent shear stress vs. apparent shear rate plot is used to identify the melt fracture phenomena. According to Ramamurthy in Journal of Rheology, 30(2), 337-357, 1986, above a certain critical flow rate, the observed extrudate irregularities may be broadly classified into two main types: surface melt fracture and gross melt fracture.

Surface melt fracture occurs under apparently steady flow conditions and ranges in detail from loss of specular gloss to the more severe form of "sharkskin". In this disclosure, the onset of surface melt fracture (OSMF) is characterized at the beginning of losing extrudate gloss at which the surface roughness of extrudate can only be

detected by 40X magnification. The critical shear rate at onset of surface melt fracture for the substantially linear ethylene/ $\alpha$ -olefin interpolymers is at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear ethylene/ $\alpha$ -olefin polymer having about the same  $I_2$  and  $M_w/M_n$ , wherein "about the same" as  
5 used herein means that each value is within 10 percent of the comparative value of the comparative linear ethylene polymer.

Gross melt fracture occurs at unsteady flow conditions and ranges in detail from regular (alternating rough and smooth, helical, etc.) to random distortions. For commercial acceptability, (for example, in blown film products), surface defects should  
10 be minimal, if not absent. The critical shear rate at onset of surface melt fracture (OSMF) and onset of gross melt fracture (OGMF) will be used herein based on the changes of surface roughness and configurations of the extrudates extruded by a GER.

The substantially linear ethylene/ $\alpha$ -olefin polymers useful for forming the compositions described herein have homogeneous branching distributions. That is, the  
15 polymers are those in which the comonomer is randomly distributed within a given interpolymers molecule and wherein substantially all of the interpolymers molecules have the same ethylene/comonomer ratio within that interpolymers. The homogeneity of the polymers is typically described by the SCBDI (Short Chain Branch Distribution Index) or CDBI (Composition Distribution Branch Index) and is defined as the weight percent  
20 of the polymer molecules having a comonomer content within 50 percent of the median total molar comonomer content. The CDBI of a polymer is readily calculated from data obtained from techniques known in the art, such as, for example, temperature rising elution fractionation (abbreviated herein as "TREF") as described, for example, in Wild et al, Journal of Polymer Science, Poly. Phys. Ed., Vol. 20, p. 441 (1982), in U.S.  
25 Patent 4,798,081 (Hazlitt et al.), or as is described in USP 5,008,204 (Stehling). The technique for calculating CDBI is described in USP 5,322,728 (Davey et al. ) and in USP 5,246,783 (Spenadel et al.). or in U.S. Patent 5,089,321 (Chum et al.). The SCBDI or CDBI for the substantially linear olefin interpolymers used in the present invention is preferably greater than 30 percent, especially greater than 50 percent. The  
30 substantially linear ethylene/ $\alpha$ -olefin interpolymers used in this invention essentially

lack a measurable "high density" fraction as measured by the TREF technique (that is, the homogeneous ethylene/ $\alpha$ -olefin interpolymers do not contain a polymer fraction with a degree of branching less than or equal to 2 methyls/1000 carbons). The substantially linear ethylene/ $\alpha$ -olefin polymers also do not contain any highly short  
5 chain branched fraction (that is, they do not contain a polymer fraction with a degree of branching equal to or more than 30 methyls/1000 carbons).

The catalysts used to prepare the homogeneous interpolymers for use as blend components in the present invention are metallocene catalysts. These metallocene catalysts include the bis(cyclopentadienyl)-catalyst systems and the  
10 mono(cyclopentadienyl) Constrained Geometry catalyst systems (used to prepare the substantially linear ethylene/ $\alpha$ -olefin polymers). Such constrained geometry metal complexes and methods for their preparation are disclosed in U.S. Application Serial No. 545,403, filed July 3, 1990 (EP-A-416,815); U.S. Application Serial No. 547,718, filed July 3, 1990 (EP-A-468,651); U.S. Application Serial No. 702,475, filed May 20,  
15 1991 (EP-A-514,828); U.S. Application Serial No. 876,268, filed May 1, 1992, (EP-A-520,732); U.S. Application Serial No. 8,003, filed January 21, 1993 (WO93/19104); U.S. Application Serial No. 08/241,523, (WO95/00526); as well as US-A-5,055,438, US-A-5,057,475, US-A-5,096,867, US-A-5,064,802, and US-A-5,132,380.

In EP-A 418,044, published March 20, 1991 (equivalent to U.S. Serial No.  
20 07/758,654) and in U.S. Serial No. 07/758,660 certain cationic derivatives of the foregoing constrained geometry catalysts that are highly useful as olefin polymerization catalysts are disclosed and claimed. In U.S. Serial Number 720,041, filed June 24, 1991, certain reaction products of the foregoing constrained geometry catalysts with various boranes are disclosed and a method for their preparation taught and claimed. In  
25 US-A 5,453,410 combinations of cationic constrained geometry catalysts with an alumoxane were disclosed as suitable olefin polymerization catalysts.

The homogeneous polymer component can be an  $\alpha$ -olefin homopolymer preferably polyethylene or polypropylene, or, preferably, an interpolymers of ethylene with at least one C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin and/or C<sub>4</sub>-C<sub>18</sub> diolefins. Homogeneous copolymers  
30 of ethylene and 1-octene are especially preferred.

### The Thermoplastic Olefins

Thermoplastic olefins (TPOs) are generally produced from polypropylene homopolymers or copolymers, or blends of an elastomeric material such as ethylene/propylene rubber (EPM) or ethylene/propylene diene monomer terpolymer (EPDM) and a more rigid material such as isotactic polypropylene. Other materials or components can be added into the formulation depending upon the application, including oil, fillers, and cross-linking agents. Generally, TPOs are characterized by a balance of stiffness (modulus) and low temperature impact, good chemical resistance and broad use temperatures. Because of features such as these, TPOs are used in many applications, including automotive facia and instrument panels, and also potentially in wire and cable

The polypropylene is generally in the isotactic form of homopolymer polypropylene, although other forms of polypropylene can also be used (for example, syndiotactic or atactic). Polypropylene impact copolymers (for example, those wherein a secondary copolymerization step reacting ethylene with the propylene is employed) and random copolymers (also reactor modified and usually containing 1.5-7percent ethylene copolymerized with the propylene), however, can also be used in the TPO formulations disclosed herein. In-reactor TPO's can also be used as blend components of the present invention. A complete discussion of various polypropylene polymers is contained in Modern Plastics Encyclopedia/89, mid October 1988 Issue, Volume 65, Number 11, pp. 86-92. The molecular weight of the polypropylene for use in the present invention is conveniently indicated using a melt flow measurement according to ASTM D-1238, Condition 230°C/2.16 kg (formerly known as "Condition (L)" and also known as I<sub>2</sub>). Melt flow rate is inversely proportional to the molecular weight of the polymer. Thus, the higher the molecular weight, the lower the melt flow rate, although the relationship is not linear. The melt flow rate for the polypropylene useful herein is generally from 0.1 grams/10 minutes (g/10 min) to 70 g/10 min, preferably from 0.5 g/10 min to 50 g/10 min, and especially from 1 g/10 min to 40 g/10 min.

### The Styrenic Block Copolymers

Also included are block copolymers having unsaturated rubber monomer units including, but not limited to, styrene-butadiene (SB), styrene-isoprene(SI), styrene-butadiene-styrene (SBS), styrene-isoprene-styrene (SIS),  $\alpha$ -methylstyrene-butadiene- $\alpha$ -methylstyrene and  $\alpha$ -methylstyrene-isoprene- $\alpha$ -methylstyrene.

The styrenic portion of the block copolymer is preferably a polymer or interpolymer of styrene and its analogs and homologs including  $\alpha$ -methylstyrene and ring-substituted styrenes, particularly ring-methylated styrenes. The preferred styrenics are styrene and  $\alpha$ -methylstyrene, and styrene is particularly preferred.

Block copolymers with unsaturated rubber monomer units may comprise homopolymers of butadiene or isoprene or they may comprise copolymers of one or both of these two dienes with a minor amount of styrenic monomer.

Preferred block copolymers with saturated rubber monomer units comprise at least one segment of a styrenic unit and at least one segment of an ethylene-butene or ethylene-propylene copolymer. Preferred examples of such block copolymers with saturated rubber monomer units include styrene/ethylene-butene copolymers, styrene/ethylene-propylene copolymers, styrene/ethylene-butene/styrene (SEBS) copolymers, styrene/ethylene-propylene/styrene (SEPS) copolymers.

### The Styrenic Copolymers

In addition to the block copolymers are the acrylonitrile-butadiene-styrene (ABS) polymers, styrene-acrylonitrile (SAN), and rubber modified styrenics including high impact polystyrene,

### The Elastomers

The elastomers include, but are not limited to, rubbers such as polyisoprene, polybutadiene, natural rubbers, ethylene/propylene rubbers, ethylene/propylene diene (EPDM) rubbers, styrene/butadiene rubbers, thermoplastic polyurethanes.

## The Thermoset Polymers

The thermoset polymers include but are not limited to epoxies, vinyl ester resins, polyurethanes, and phenolics.

## 5 The Vinyl Halide Polymers

Vinyl halide homopolymers and copolymers are a group of resins which use as a building block the vinyl structure  $\text{CH}_2=\text{CXY}$ , where X is selected from the group consisting of F, Cl, Br, and I and Y is selected from the group consisting of F, Cl, Br, I and H.

10 The vinyl halide polymer component of the blends of the present invention include but are not limited to homopolymers and copolymers of vinyl halides with copolymerizable monomers such as  $\alpha$ -olefins including but not limited to ethylene, propylene, vinyl esters of organic acids containing 1 to 18 carbon atoms, for example vinyl acetate, vinyl stearate and so forth; vinyl chloride, vinylidene chloride,  
15 symmetrical dichloroethylene; acrylonitrile, methacrylonitrile; alkyl acrylate esters in which the alkyl group contains 1 to 8 carbon atoms, for example methyl acrylate and butyl acrylate; the corresponding alkyl methacrylate esters; dialkyl esters of dibasic organic acids in which the alkyl groups contain 1 - 8 carbon atoms, for example dibutyl fumarate, diethyl maleate, and so forth.

20 Preferably the vinyl halide polymers are homopolymers or copolymers of vinyl chloride or vinylidene chloride. Poly (vinyl chloride) polymers (PVC) can be further classified into two main types by their degree of rigidity. These are "rigid" PVC and "flexible" PVC. Flexible PVC is distinguished from rigid PVC primarily by the presence of and amount of plasticizers in the resin. Flexible PVC typically has  
25 improved processability, lower tensile strength and higher elongation than rigid PVC.

Of the vinylidene chloride homopolymers and copolymers (PVDC), typically the copolymers with vinyl chloride, acrylates or nitriles are used commercially and are most preferred. The choice of the comonomer significantly affects the properties of the resulting polymer. Perhaps the most notable properties of the various PVDC's are their  
30 low permeability to gases and liquids, barrier properties; and chemical resistance.

Also included are the various PVC and PVCD formulations containing minor amounts of other materials present to modify the properties of the PVC or PVCD, including but not limited to polystyrene, styrenic copolymers, polyolefins including homo and copolymers comprising polyethylene, and or polypropylene, and other  
5 ethylene/ $\alpha$ -olefin copolymers, polyacrylic resins, butadiene-containing polymers such as acrylonitrile butadiene styrene terpolymers (ABS), and methacrylate butadiene styrene terpolymers (MBS), and chlorinated polyethylene (CPE) resins.

Also included in the family of vinyl halide polymers for use as blend components of the present invention are the chlorinated derivatives of PVC typically  
10 prepared by post chlorination of the base resin and known as chlorinated PVC, (CPVC). Although CPVC is based on PVC and shares some of its characteristic properties, CPVC is a unique polymer having a much higher melt temperature range (410 - 450°C) and a higher glass transition temperature (239 - 275°F) than PVC.

#### 15 Tackifiers

Tackifiers can also be added to the polymer compositions used to prepare the fibers of the present invention in order to further increase the Tg and thus extend the application temperature window of the fibers, fabrics and fabricated articles therefrom.

A suitable tackifier may be selected on the basis of the criteria outlined by  
20 Hercules in J. Simons, Adhesives Age, "The HMDA Concept: A New Method for Selection of Resins", November 1996. This reference discusses the importance of the polarity and molecular weight of the resin in determining compatibility with the polymer.

In the case of substantially random interpolymers of at least one  $\alpha$ -olefin and a  
25 vinyl aromatic monomer, preferred tackifiers will have some degree of aromatic character to promote compatibility, particularly in the case of substantially random interpolymers having a high content of the vinyl aromatic monomer. As an initial indicator, compatible tackifiers are those which are also known to be compatible with ethylene/vinyl acetate having 28 weight percent vinyl acetate.

Tackifying resins can be obtained by the polymerization of petroleum and terpene feedstreams and from the derivatization of wood, gum, and tall oil rosin. Several classes of tackifiers include wood rosin, tall oil and tall oil derivatives, and cyclopentadiene derivatives, such as are described in United Kingdom patent application GB 2,032,439A. Other classes of tackifiers include aliphatic C<sub>5</sub> resins, polyterpene resins, hydrogenated resins, mixed aliphatic-aromatic resins, rosin esters, natural and synthetic terpenes, terpene-phenolics, and hydrogenated rosin esters.

Rosin is a commercially available material that occurs naturally in the oleo rosin of pine trees and typically is derived from the oleo resinous exudate of the living tree, from aged stumps and from tall oil produced as a by-product of kraft paper manufacture. After it is obtained, rosin can be treated by hydrogenation, dehydrogenation, polymerization, esterification, and other post treatment processes. Rosin is typically classed as a gum rosin, a wood rosin, or as a tall oil rosin which indicate its source. The materials can be used unmodified, in the form of esters of polyhydric alcohols, and can be polymerized through the inherent unsaturation of the molecules. These materials are commercially available and can be blended into the compositions using standard blending techniques. Representative examples of such rosin derivatives include pentaerythritol esters of tall oil, gum rosin, wood rosin, or mixtures thereof.

Examples of the various classes of tackifiers include, but are not limited to, aliphatic resins, polyterpene resins, hydrogenated resins, mixed aliphatic-aromatic resins, styrene/ $\alpha$ -methylene styrene resins, pure monomer hydrocarbon resin, hydrogenated pure monomer hydrocarbon resin, modified styrene copolymers, pure aromatic monomer copolymers, and hydrogenated aliphatic hydrocarbon resins.

Exemplary aliphatic resins include those available under the trade designations Escorez<sup>TM</sup>, Piccotac<sup>TM</sup>, Mercures<sup>TM</sup>, Wingtack<sup>TM</sup>, Hi-Rez<sup>TM</sup>, Quintone<sup>TM</sup>, Tackirol<sup>TM</sup>, etc. Exemplary polyterpene resins include those available under the trade designations Nirez<sup>TM</sup>, Piccolyte<sup>TM</sup>, Wingtack<sup>TM</sup>, Zonarez<sup>TM</sup>, etc. Exemplary hydrogenated resins include those available under the trade designations Escorez<sup>TM</sup>, Arkon<sup>TM</sup>, Clearon<sup>TM</sup>, etc. Exemplary mixed aliphatic-aromatic resins include those available under the trade



designations Escorez™, Regalite™, Hercures™, AR™, Imprez™, Norsolene™ M, Marukarez™, Arkon™ M, Quintone™, Wingtack™, etc. One particularly preferred class of tackifiers includes the styrene/α-methylene styrene tackifiers available from Hercules. Particularly suitable classes of tackifiers include Wingtack™ 86 and  
5 Hercotac™ 1149, Eastman H-130, and styrene/α-methyl styrene tackifiers. Other preferred tackifiers include Piccotex 75, a pure monomer hydrocarbon resin having a glass transition temperature of 33°C, available from Hercules, Regalrez™ 1139 which is prepared by polymerization and hydrogenation of pure monomer hydrocarbon, Picotex™ 120 which is a copolymer of modified styrene, Kristalex™ 5140 which is a  
10 copolymer of the pure aromatic monomers, Plastolyn™ 140 which is a hydrogenated aliphatic hydrocarbon resin, and Endex™ 155 which is a copolymer of the pure aromatic monomers. Of these Kristalex™ 5140, Plastolyn™ 140, and Endex™ 155 are preferred and Endex™ 155 is most preferred.

#### 15 Other Additives

Additives such as antioxidants (for example, hindered phenols such as, for example, Irganox® 1010), phosphites (for example, Irgafos® 168), u.v. stabilizers, cling additives (for example, polyisobutylene), antiblock additives, colorants, pigments, slip agents (e.g. stearamide and/or erucamide) can also be included in the interpolymers  
20 and/or blends employed to prepare the fibers of the present invention, to the extent that they do not interfere with the properties of the substantially random interpolymers. Processing aids, which are also referred to herein as plasticizers, are optionally provided to reduce the viscosity of a composition, and include the phthalates, such as dioctyl phthalate and diisobutyl phthalate, natural oils such as lanolin, and paraffin,  
25 naphthenic and aromatic oils obtained from petroleum refining, and liquid resins from rosin or petroleum feedstocks. Exemplary classes of oils useful as processing aids include white mineral oil (such as Kaydol™ oil (available from Witco), and Shellflex™ 371 naphthenic oil (available from Shell Oil Company). Another suitable oil is Tuflo™ oil (available from Lyondell).

Also included as a potential component of the polymer compositions used in the present invention are various organic and inorganic fillers, the identity of which depends upon the type of application in the blend is to be utilized.). Representative examples of such fillers include organic and inorganic fibers such as those made from  
5 asbestos, boron, graphite, ceramic, glass, metals (such as stainless steel) or polymers (such as aramid fibers) talc, carbon black, carbon fibers, calcium carbonate, alumina trihydrate, glass fibers, marble dust, cement dust, clay, feldspar, silica or glass, fumed silica, alumina, magnesium oxide, magnesium hydroxide, antimony oxide, zinc oxide, barium sulfate, aluminum silicate, calcium silicate, titanium dioxide, titanates,  
10 aluminum nitride,  $B_2O_3$ , nickel powder or chalk.

Other representative organic or inorganic, fiber or mineral, fillers include carbonates such as barium, calcium or magnesium carbonate; fluorides such as calcium or sodium aluminum fluoride; hydroxides such as aluminum hydroxide; metals such as aluminum, bronze, lead or zinc; oxides such as aluminum, antimony, magnesium or  
15 zinc oxide, or silicon or titanium dioxide; silicates such as asbestos, mica, clay (kaolin or calcined kaolin), calcium silicate, feldspar, glass (ground or flaked glass or hollow glass spheres or microspheres or beads, whiskers or filaments), nepheline, perlite, pyrophyllite, talc or wollastonite; sulfates such as barium or calcium sulfate; metal sulfides; cellulose, in forms such as wood or shell flour; calcium terephthalate; and  
20 liquid crystals. Mixtures of more than one such filler may be used as well.

These additives are employed in functionally equivalent amounts known to those skilled in the art. For example, the amount of antioxidant employed is that amount which prevents the polymer or polymer blend from undergoing oxidation at the temperatures and environment employed during storage and ultimate use of the  
25 polymers. Such amount of antioxidants is usually in the range of from 0.01 to 10, preferably from 0.05 to 5, more preferably from 0.1 to 2 percent by weight based upon the weight of the polymer or polymer blend. Similarly, the amounts of any of the other enumerated additives are the functionally equivalent amounts such as the amount to render the polymer or polymer blend antiblocking, to produce the desired result, to  
30 provide the desired color from the colorant or pigment. Such additives can suitably be

employed in the range of from 0.05 to 50, preferably from 0.1 to 35, more preferably from 0.2 to 20 percent by weight based upon the weight of the polymer or polymer blend. When a processing aid is employed, it will be present in the composition of the invention in an amount of at least 5 percent. The processing aid will typically be  
5 present in an amount of no more than 60, preferably no more than 30, and most preferably no more than 20 weight percent.

#### Preparation of the Blends Comprising the Substantially Random Interpolymers

The blended polymer compositions used to prepare the fabricated articles of the  
10 present invention can be prepared by any convenient method, including dry blending the individual components and subsequently melt mixing or melt compounding in a Haake torque rheometer or, either directly in the extruder or mill used to make the finished article (for example, the automotive part), or by pre-melt mixing in a separate extruder or mill (for example, a Banbury mixer), or by solution blending, or by  
15 compression molding, or by calendering.

#### Properties of the Fibers and/or Fabric of the Present Invention

Various homofil fibers can be made from the substantially random  
interpolymers. The shape of the fiber is not limited. For example, typical fiber have a  
20 circular cross sectional shape, but sometimes fibers have different shapes, such as a trilobal shape, or a flat (that is, "ribbon" like) shape to promote ease of handling. The fiber disclosed herein is not limited by the shape of the fiber.

For the novel fibers disclosed herein, the diameter can be widely varied. However, the fiber denier can be adjusted to suit the capabilities of the finished article  
25 and as such, would preferably be: from 0.5 to 30 denier/filament for melt blown; from 1 to 30 denier/filament for spunbond; and from 1 to 20,000 denier/filament for continuous wound filament.

The polymer compositions used to prepare the fibers of the present invention comprise from 1 to 100, preferably from 10 to 100, more preferably from 50 to 100,  
30 even more preferably from 80 to 100 wt percent, (based on the combined weights of

this component and the polymer component other than the substantially random interpolymers) of one or more interpolymers of one or more  $\alpha$ -olefins and one or more vinyl or aromatic monomers and/or one or more hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers.

5           The substantially random interpolymers can be used as a minor component of a multi-component blend when used as for example, a compatibilizer or bonding component, it can be present in amounts even more preferably from 80 to 100 wtpercent, (based on the combined weights of this component and the polymer component other than the substantially random interpolymers).

10           For the polymer compositions used to prepare the fibers of the present invention comprising only the substantially random interpolymers and a tackifier, the substantially random interpolymers can be present in amounts from 50 to 100, preferably from 50 to 95, more preferably from 60 to 90 wtpercent, (based on the combined weights of this component and the tackifier).

15           The substantially random interpolymers usually contain from 0.5 to 65 preferably from 1 to 55, more preferably from 2 to 50 mole percent of at least one vinyl or vinylidene aromatic monomer and/or hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer and from 35 to 99.5, preferably from 45 to 99, more preferably from 50 to 98 mole percent of at least one aliphatic  $\alpha$ -olefin having from 2 to 20 carbon  
20 atoms.

          The number average molecular weight ( $M_n$ ) of the substantially random interpolymers used to prepare the fibers of the present invention is greater than 1000, preferably from 5,000 to 1,000,000, more preferably from 10,000 to 500,000.

          The melt index ( $I_2$ ) of the substantially random interpolymers used to prepare the  
25 fibers of the present invention is from 0.1 to 1,000, preferably of from 0.5 to 200, more preferably of from 0.5 to 100 g/10 min.

          The molecular weight distribution ( $M_w/M_n$ ) of the substantially random interpolymers used to prepare the fibers of the present invention is from 1.5 to 20, preferably of from 1.8 to 10, more preferably of from 2 to 5.

The density of the substantially random interpolmer used to prepare the fibers of the present invention is greater than 0.930, preferably from 0.930 to 1.045, more preferably of from 0.930 to 1.040, most preferably of from 0.930 to 1.030 g/cm<sup>3</sup>.

The polymer compositions used to prepare the homofil fibers of the present invention can also comprise from 0 to 99, preferably from 0 to 90, more preferably from 0 to 50, even more preferably 0 to 20 percent of by weight of at least one polymer other than the substantially random interpolmer (based on the combined weights of this component and the substantially random interpolmer) which can comprise a homogenous  $\alpha$ -olefin homopolymer or interpolmer comprising polypropylene, propylene/C<sub>4</sub>-C<sub>20</sub>  $\alpha$ -olefin copolymers, polyethylene, and ethylene/C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin copolymers, the interpolymers can be either heterogeneous ethylene/ $\alpha$ -olefin interpolymers, preferably a heterogeneous ethylene/C<sub>3</sub>-C<sub>8</sub>  $\alpha$ -olefin interpolmer, most preferably a heterogeneous ethylene/octene-1 interpolmer or homogeneous ethylene/ $\alpha$ -olefin interpolymers, including the substantially linear ethylene/ $\alpha$ -olefin interpolymers, preferably a substantially linear ethylene/ $\alpha$ -olefin interpolmer, most preferably a substantially linear ethylene/C<sub>3</sub>-C<sub>8</sub>  $\alpha$ -olefin interpolmer; or a heterogeneous ethylene/ $\alpha$ -olefin interpolmer; or a thermoplastic olefin, preferably an ethylene/propylene rubber (EPM) or ethylene/propylene diene monomer terpolymer (EPDM) or isotactic polypropylene, most preferably isotactic polypropylene; or a styrenic block copolymer, preferably styrene-butadiene (SB), styrene-isoprene (SI), styrene-butadiene-styrene (SBS), styrene-isoprene-styrene (SIS) or styrene-ethylene/butene-styrene (SEBS) block copolymer, most preferably a styrene-butadiene-styrene (SBS) copolymer; or styrenic homopolymers or copolymers, preferably polystyrene, high impact polystyrene, polyvinyl chloride, copolymers of styrene and at least one of acrylonitrile, meth-acrylonitrile, maleic anhydride, or  $\alpha$ -methyl styrene, most preferably polystyrene, or elastomers, preferably polyisoprene, polybutadiene, natural rubbers, ethylene/propylene rubbers, ethylene/propylene diene (EPDM) rubbers, styrene/butadiene rubbers, thermoplastic polyurethanes, most preferably thermoplastic polyurethanes; or thermoset polymers, preferably epoxies, vinyl ester resins, polyurethanes, phenolics, most preferably polyurethanes; or vinyl halide homopolymers

and copolymers, preferably homopolymers or copolymers of vinyl chloride or vinylidene chloride or the chlorinated derivatives therefrom, most preferably poly (vinyl chloride) and poly (vinylidene chloride); or engineering thermoplastics, preferably poly(methylmethacrylate) (PMMA), cellulotics, nylons, poly(esters),  
5 poly(acetals); poly(amides), the poly(arylate), aromatic polyesters, poly(carbonate), poly(butylene) and polybutylene and polyethylene terephthalates, most preferably poly(methylmethacrylate) (PMMA), and poly(esters).

The polymer composition used to prepare the fibers of the present invention can also comprise from 0 to 50, preferably from 5 to 50, more preferably from 10 to 40  
10 percent by weight (based on the final weight of the polymer or polymer blend) of one or more tackifiers comprising aliphatic resins, polyterpene resins, hydrogenated resins, mixed aliphatic-aromatic resins, styrene/ $\alpha$ -methylene styrene resins, pure monomer hydrocarbon resin, hydrogenated pure monomer hydrocarbon resin, modified styrene copolymers, pure aromatic monomer copolymers, and hydrogenated aliphatic  
15 hydrocarbon resins.

For the bicomponent fibers of the present invention the first component comprises a substantially random interpolmer having the compositions and properties as used to prepare the homofil fibers of the present invention and present in an amount of from 5 to 95, preferably from 25 to 95, most preferably from 50 to 95 wt percent  
20 (based on the combined weight of the first and second components of the bicomponent fiber). The second component is present in an amount of from 5 to 95, preferably from 5 to 75, most preferably from 5 to 50 wt percent (based on the combined weight of the first and second components of the bicomponent fiber).

The following examples are illustrative of the invention, but are not to be  
25 construed as to limiting the scope thereof in any manner.

## EXAMPLES

Test Methods.a) Melt Flow and Density Measurements

5           The molecular weight of the polymer compositions for use in the present invention is conveniently indicated using a melt index measurement according to ASTM D-1238, Condition 190°C/2.16 kg (formally known as "Condition (E)" and also known as I<sub>2</sub>) was determined. Melt index is inversely proportional to the molecular weight of the polymer. Thus, the higher the molecular weight, the lower the melt  
10   index, although the relationship is not linear.

          Also useful for indicating the molecular weight of the substantially random interpolymers used in the present invention is the Gottfert melt index (G, cm<sup>3</sup>/10 min) which is obtained in a similar fashion as for melt index (I<sub>2</sub>) using the ASTM D1238 procedure for automated plastometers, with the melt density set to 0.7632, the melt  
15   density of polyethylene at 190 deg. C.

          The relationship of melt density to styrene content for ethylene-styrene interpolymers was measured, as a function of total styrene content, at 190° C for a range of 29.8percent to 81.8percent by weight styrene . Atactic polystyrene levels in these samples was typically 10percent or less. The influence of the atactic polystyrene  
20   was assumed to be minimal because of the low levels. Also, the melt density of atactic polystyrene and the melt densities of the samples with high total styrene are very similar.

          The method used to determine the melt density employed a Gottfert melt index machine with a melt density parameter set to 0.7632, and the collection of melt strands  
25   as a function of time while the I<sub>2</sub> weight was in force. The weight and time for each melt strand was recorded and normalized to yield the mass in grams per 10 minutes. The instrument's calculated I<sub>2</sub> melt index value was also recorded. The equation used to calculate the actual melt density is

$$\delta = \delta_{0.7632} \times I_2 / I_2 \text{ Gottfert}$$

30   where  $\delta_{0.7632} = 0.7632$  and I<sub>2</sub> Gottfert = displayed melt index.

A linear least squares fit of calculated melt density versus total styrene content leads to an equation with a correlation coefficient of 0.91 for the following equation:

$$\delta = 0.00299 \times S + 0.723$$

where  $S$  = weight percentage of styrene in the polymer. The relationship of total styrene to melt density can be used to determine an actual melt index value, using these equations if the styrene content is known.

So for a polymer that is 73percent total styrene content with a measured melt flow (the "Gottfert number"), the calculation becomes:

$$x = 0.00299 \times 73 + 0.723 = 0.9412$$

10

where

$$0.9412/0.7632 = I_2 / G\# \text{ (measured)} = 1.23$$

The density of the substantially random interpolymers used in the present invention was determined in accordance with ASTM D-792.

#### 15 b) Styrene Analyses

Interpolymer styrene content and atactic polystyrene concentration were determined using proton nuclear magnetic resonance ( $^1\text{H}$  N.M.R). All proton NMR samples were prepared in 1, 1, 2, 2-tetrachloroethane- $\text{d}_2$  (TCE- $\text{d}_2$ ). The resulting solutions were 1.6 - 3.2 percent polymer by weight. Melt index ( $I_2$ ) was used as a guide for determining sample concentration. Thus when the  $I_2$  was greater than 2 g/10 min, 40 mg of interpolymer was used; with an  $I_2$  between 1.5 and 2 g/10 min, 30 mg of interpolymer was used; and when the  $I_2$  was less than 1.5 g/10 min, 20 mg of interpolymer was used. The interpolymers were weighed directly into 5 mm sample tubes. A 0.75 mL aliquot of TCE- $\text{d}_2$  was added by syringe and the tube was capped with a tight-fitting polyethylene cap. The samples were heated in a water bath at 85°C to soften the interpolymer. To provide mixing, the capped samples were occasionally brought to reflux using a heat gun.

Proton NMR spectra were accumulated on a Varian VXR 300 with the sample probe at 80°C, and referenced to the residual protons of TCE- $\text{d}_2$  at 5.99 ppm. The delay times were varied between 1 second, and data was collected in triplicate on each



sample. The following instrumental conditions were used for analysis of the interpolmer samples:

Varian VXR-300, standard  $^1\text{H}$ :

Sweep Width, 5000 Hz

5 Acquisition Time, 3.002 sec

Pulse Width, 8  $\mu\text{sec}$

Frequency, 300 MHz

Delay, 1 sec

Transients, 16

10 The total analysis time per sample was about 10 minutes.

Initially, a  $^1\text{H}$  NMR spectrum for a sample of the polystyrene, Styron<sup>TM</sup> 680 (available from the Dow Chemical Company, Midland, MI) was acquired with a delay time of one second. The protons were "labeled": b, branch;  $\alpha$ , alpha; o, ortho; m, meta; p, para, as shown in Figure 1.

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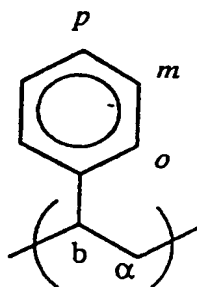


Figure 1.

Integrals were measured around the protons labeled in Figure 1; the 'A' designates aPS. Integral  $A_{7.1}$  (aromatic, around 7.1 ppm) is believed to be the three ortho/para protons; and integral  $A_{6.6}$  (aromatic, around 6.6 ppm) the two meta protons. The two aliphatic protons labeled  $\alpha$  resonate at 1.5 ppm; and the single proton labeled b is at 1.9 ppm. The aliphatic region was integrated from about 0.8 to 2.5 ppm and is referred to as  $A_{al}$ . The theoretical ratio for  $A_{7.1}$ :  $A_{6.6}$ :  $A_{al}$  is 3: 2: 3, or 1.5: 1: 1.5, and correlated very well with the observed ratios for the Styron<sup>TM</sup> 680 sample for several delay times of 1 second. The ratio calculations used to check the integration and verify

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peak assignments were performed by dividing the appropriate integral by the integral  $A_{6.6}$ . Ratio  $A_r$  is  $A_{7.1} / A_{6.6}$ .

Region  $A_{6.6}$  was assigned the value of 1. Ratio  $A_l$  is integral  $A_{al} / A_{6.6}$ . All spectra collected have the expected 1.5: 1: 1.5 integration ratio of (*o+p*): *m*: ( $\alpha+b$ ). The ratio of aromatic to aliphatic protons is 5 to 3. An aliphatic ratio of 2 to 1 is predicted based on the protons labeled  $\alpha$  and  $b$  respectively in Figure 1. This ratio was also observed when the two aliphatic peaks were integrated separately.

For the ethylene/styrene interpolymers, the  $^1\text{H}$  NMR spectra using a delay time of one second, had integrals  $C_{7.1}$ ,  $C_{6.6}$ , and  $C_{al}$  defined, such that the integration of the peak at 7.1 ppm included all the aromatic protons of the copolymer as well as the *o* & *p* protons of aPS. Likewise, integration of the aliphatic region  $C_{al}$  in the spectrum of the interpolymers included aliphatic protons from both the aPS and the interpolymers with no clear baseline resolved signal from either polymer. The integral of the peak at 6.6 ppm  $C_{6.6}$  is resolved from the other aromatic signals and it is believed to be due solely to the aPS homopolymer (probably the *meta* protons). (The peak assignment for atactic polystyrene at 6.6 ppm (integral  $A_{6.6}$ ) was made based upon comparison to the authentic sample Styron<sup>TM</sup> 680.) This is a reasonable assumption since, at very low levels of atactic polystyrene, only a very weak signal is observed here. Therefore, the phenyl protons of the copolymer must not contribute to this signal. With this assumption, integral  $A_{6.6}$  becomes the basis for quantitatively determining the aPS content.

The following equations were then used to determine the degree of styrene incorporation in the ethylene/styrene interpolymers samples:

$$(C \text{ Phenyl}) = C_{7.1} + A_{7.1} - (1.5 \times A_{6.6})$$

$$(C \text{ Aliphatic}) = C_{al} - (1.5 \times A_{6.6})$$

$$s_c = (C \text{ Phenyl}) / 5$$

$$e_c = (C \text{ Aliphatic} - (3 \times s_c)) / 4$$

$$E = e_c / (e_c + s_c)$$

$$S_c = s_c / (e_c + s_c)$$

and the following equations were used to calculate the mol percent ethylene and styrene in the interpolymers.

$$\text{Wt \% E} = \frac{E * 28}{(E * 28) + (S_c * 104)} (100)$$

and

$$\text{Wt \% S} = \frac{S_c * 104}{(E * 28) + (S_c * 104)} (100)$$

5

where:  $s_c$  and  $e_c$  are styrene and ethylene proton fractions in the interpolymer, respectively, and  $S_c$  and  $E$  are mole fractions of styrene monomer and ethylene monomer in the interpolymer, respectively.

The weight percent of aPS in the interpolymers was then determined by the  
10 following equation:

$$\text{Wt \% aPS} = \frac{(\text{Wt \% S}) * \left( \frac{A_{6.6/2}}{S_c} \right)}{100 + \left[ (\text{Wt \% S}) * \left( \frac{A_{6.6/2}}{S_c} \right) \right]} * 100$$

The total styrene content was also determined by quantitative Fourier Transform Infrared spectroscopy (FTIR).

Test parts and characterization data for the interpolymers and their blends are  
15 generated according to the following procedures:

#### Compression Molding.

Samples are melted at 190°C for 3 minutes and compression molded at 190°C under 20,000 lb (9,072 kg) of pressure for another 2 minutes. Subsequently, the molten materials are quenched in a press equilibrated at room temperature.

#### 20 Injection Molding.

Samples were injection molded on a 150 ton deMag injection molding machine at 190 C melt temperature, 1 second injection time, 70 F water temperature, and 60 second overall cycle time. The mold was an ASTM test mold which includes 0.5 inch by 5 inch by 75 mil thick ASTM flexural modulus test specimens.

Differential Scanning Calorimetry (DSC).

A Dupont DSC-2920 is used to measure the thermal transition temperatures and heat of transition for the interpolymers. In order to eliminate previous thermal history, samples are first heated to 200°C. Heating and cooling curves are recorded at  
5 10°C/min. Melting (from second heat) and crystallization temperatures are recorded from the peak temperatures of the endotherm and exotherm, respectively.

Preparation of ESI Interpolymers Used in Examples and Comparative Experiments of Present Invention10 1) Preparation of ESI #'s 1-6

The interpolymers were prepared in a 400 gallon(1514 L) agitated semi-continuous batch reactor. The reaction mixture consisted of approximately 250 gallons (946 L) of solvent comprising a mixture of cyclohexane (85 wt percent) and isopentane (15 wt percent), and styrene. Prior to addition, solvent, styrene and ethylene were purified to  
15 remove water and oxygen. The inhibitor in the styrene was also removed. Inerts were removed by purging the vessel with ethylene. The vessel was then pressure controlled to a set point with ethylene. Hydrogen was added to control molecular weight. Temperature in the vessel was controlled to set-point by varying the jacket water temperature on the vessel. Prior to polymerization, the vessel was heated to the desired  
20 run temperature and the catalyst components Titanium: (N-1,1-dimethylethyl)dimethyl(1-(1,2,3,4,5-eta)-2,3,4,5-tetramethyl- 2,4-cyclopentadien-1-yl)silanaminate))(2-)N)-dimethyl, CAS# 135072-62-7 and Tris(pentafluorophenyl)boron, CAS# 001109-15-5, Modified methylaluminoxane Type 3A, CAS# 146905-79-5 were flow controlled, on a mole ratio basis of 1/3/5  
25 respectively , combined and added to the vessel. After starting, the polymerization was allowed to proceed with ethylene supplied to the reactor as required to maintain vessel pressure. In some cases, hydrogen was added to the headspace of the reactor to maintain a mole ratio with respect to the ethylene concentration. At the end of the run, the catalyst flow was stopped, ethylene was removed from the reactor, about 1000 ppm  
30 of Irganox™ 1010 anti-oxidant (trademark of Ciba Geigy Corp.) was then added to the

solution and the polymer was isolated from the solution. The resulting polymers were isolated from solution by either stripping with steam in a vessel or by use of a devolatilizing extruder. In the case of the steam stripped material, additional processing was required in extruder like equipment to reduce residual moisture and any unreacted  
5 styrene. The specific preparation conditions for each interpolymer are summarized in Table 1 and their properties in Table 2.

Table 1. Preparation Conditions for ESI #'s 1 - 6

ESI #	Solvent loaded		Styrene loaded		Pressure		Temp.	Total H <sub>2</sub> Added	Run Time
	lbs	kg	lbs	kg	Psig	kPa	° C	Grams	Hrs
ESI 1	252	114	1320	599	40	276	60	23	6.5
ESI 2	842	381	662	300	105	724	60	8.8	3.7
ESI 3	840	380	661	299	105	724	60	36.5	5.0
ESI 4	839	380	661	299	105	724	60	53.1	4.8
ESI 5	1196	541	225	102	70	483	60	7.5	6.1
ESI 6	1196	541	225	102	70	483	60	81.1	4.8

Table 2. Properties of ESI #'s 1 - 6

ESI #	ESI Styrene (wt%)	ESI Styrene (mol%)	Atactic Polystyrene (wt%)	Melt Index, I <sub>2</sub> (g/10m)	10 <sup>-3</sup> M <sub>w</sub>	M <sub>w</sub> /M <sub>n</sub> Ratio	Tg (°C)	Tensile Modulus (KPSI)	Flex Modulus (KPSI)
ESI 1	72.7	41.8	7.8	1.83	187	2.63	24.7	102	90
ESI 2	45.0	18.0	4.0	0.01	327	2.26	- 12.7	1	20
ESI 3	45.7	18.5	N/A	0.72	N/A	N/A	N/A	N/A	N/A
ESI 4	43.4	17.1	10.3	2.62	126	1.89	-4.4	1	10
ESI 5	27.3	9.2	1.2	0.03	241	2.04	- 17.2	3	9
ESI 6	32.5	11.5	7.8	10.26	83	1.87	- 15.8	3	6

## 5 2) Preparation of ESI #'s 7-31

ESI #'s 7 - 31 are substantially random ethylene/styrene interpolymers prepared using the following catalyst and polymerization procedures.

## 10 Preparation of Catalyst A (dimethyl[N-(1,1-dimethylethyl)-1,1-dimethyl-1-[(1,2,3,4,5- $\eta$ )-1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl]silanaminato(2-)-N]-titanium).

### 1) Preparation of 3,5,6,7-Tetrahydro-s-Hydrindacen-1(2H)-one.

Indan (94.00 g, 0.7954 moles) and 3-chloropropionyl chloride (100.99 g, 0.7954 moles) were stirred in  $\text{CH}_2\text{Cl}_2$  (300 mL) at  $0^\circ\text{C}$  as  $\text{AlCl}_3$  (130.00 g, 0.9750 moles) was added slowly under a nitrogen flow. The mixture was then allowed to stir at room temperature for 2 hours. The volatiles were then removed. The mixture was then cooled to  $0^\circ\text{C}$  and concentrated  $\text{H}_2\text{SO}_4$  (500 mL) slowly added. The forming solid had to be frequently broken up with a spatula as stirring was lost early in this step. The mixture was then left under nitrogen overnight at room temperature. The mixture was then heated until the temperature readings reached  $90^\circ\text{C}$ . These conditions were maintained for a 2 hour period of time during which a spatula was periodically used to stir the mixture. After the reaction period crushed ice was placed in the mixture and moved around. The mixture was then transferred to a beaker and washed intermittently with  $\text{H}_2\text{O}$  and diethyl ether and then the fractions filtered and combined. The mixture was washed with  $\text{H}_2\text{O}$  (2 x 200 mL). The organic layer was then separated and the volatiles removed. The desired product was then isolated via recrystallization from hexane at  $0^\circ\text{C}$  as pale yellow crystals (22.36 g, 16.3percent yield).

$^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$ 2.04-2.19 (m, 2 H), 2.65 (t,  $^3J_{\text{HH}}=5.7$  Hz, 2 H), 2.84-3.0 (m, 4 H), 3.03 (t,  $^3J_{\text{HH}}=5.5$  Hz, 2 H), 7.26 (s, 1 H), 7.53 (s, 1 H).

$^{13}\text{C}$  NMR ( $\text{CDCl}_3$ ):  $\delta$ 25.71, 26.01, 32.19, 33.24, 36.93, 118.90, 122.16, 135.88, 144.06, 152.89, 154.36, 206.50.

GC-MS: Calculated for  $\text{C}_{12}\text{H}_{12}\text{O}$  172.09, found 172.05.

## 2) Preparation of 1,2,3,5-Tetrahydro-7-phenyl-s-indacen.

3,5,6,7-Tetrahydro-s-Hydrindacen-1(2H)-one (12.00 g, 0.06967 moles) was stirred in diethyl ether (200 mL) at  $0^\circ\text{C}$  as  $\text{PhMgBr}$  (0.105 moles, 35.00 mL of 3.0 M solution in diethyl ether) was added slowly. This mixture was then allowed to stir overnight at room temperature. After the reaction period the mixture was quenched by pouring over ice. The mixture was then acidified ( $\text{pH}=1$ ) with  $\text{HCl}$  and stirred vigorously for 2 hours. The organic layer was then separated and washed with  $\text{H}_2\text{O}$  (2 x 100 mL) and then dried over  $\text{MgSO}_4$ . Filtration followed by the removal of the

volatiles resulted in the isolation of the desired product as a dark oil (14.68 g, 90.3percent yield).

$^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$ 2.0-2.2 (m, 2 H), 2.8-3.1 (m, 4 H), 6.54 (s, 1H), 7.2-7.6 (m, 7 H).

GC-MS: Calculated for  $\text{C}_{18}\text{H}_{16}$  232.13, found 232.05.

5

3) Preparation of 1,2,3,5-Tetrahydro-7-phenyl-s-indacene, dilithium salt.

1,2,3,5-Tetrahydro-7-phenyl-s-indacen (14.68 g, 0.06291 moles) was stirred in hexane (150 mL) as nBuLi (0.080 moles, 40.00 mL of 2.0 M solution in cyclohexane) was slowly added. This mixture was then allowed to stir overnight. After the reaction  
10 period the solid was collected via suction filtration as a yellow solid which was washed with hexane, dried under vacuum, and used without further purification or analysis (12.2075 g, 81.1percent yield).

4) Preparation of Chlorodimethyl(1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl)silane.

15 1,2,3,5-Tetrahydro-7-phenyl-s-indacene, dilithium salt (12.2075 g, 0.05102 moles) in THF (50 mL) was added dropwise to a solution of  $\text{Me}_2\text{SiCl}_2$  (19.5010 g, 0.1511 moles) in THF (100 mL) at  $0^\circ\text{C}$ . This mixture was then allowed to stir at room temperature overnight. After the reaction period the volatiles were removed and the residue extracted and filtered using hexane. The removal of the hexane resulted in the  
20 isolation of the desired product as a yellow oil (15.1492 g, 91.1percent yield).

$^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$ 0.33 (s, 3 H), 0.38 (s, 3 H), 2.20 (p,  $^3\text{J}_{\text{HH}}=7.5$  Hz, 2 H), 2.9-3.1 (m, 4 H), 3.84 (s, 1 H), 6.69 (d,  $^3\text{J}_{\text{HH}}=2.8$  Hz, 1 H), 7.3-7.6 (m, 7 H), 7.68 (d,  $^3\text{J}_{\text{HH}}=7.4$  Hz, 2 H).

$^{13}\text{C}$  NMR ( $\text{CDCl}_3$ ):  $\delta$ 0.24, 0.38, 26.28, 33.05, 33.18, 46.13, 116.42, 119.71, 127.51,  
25 128.33, 128.64, 129.56, 136.51, 141.31, 141.86, 142.17, 142.41, 144.62.

GC-MS: Calculated for  $\text{C}_{20}\text{H}_{21}\text{ClSi}$  324.11, found 324.05.

5) Preparation of N-(1,1-Dimethylethyl)-1,1-dimethyl-1-(1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl)silanamine.



Chlorodimethyl(1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl)silane (10.8277 g, 0.03322 moles) was stirred in hexane (150 mL) as NEt<sub>3</sub> (3.5123 g, 0.03471 moles) and *t*-butylamine (2.6074 g, 0.03565 moles) were added. This mixture was allowed to stir for 24 hours. After the reaction period the mixture was filtered and the volatiles removed resulting in the isolation of the desired product as a thick red-yellow oil (10.6551 g, 88.7percent yield).

<sup>1</sup>H NMR (CDCl<sub>3</sub>): δ 0.02 (s, 3 H), 0.04 (s, 3 H), 1.27 (s, 9 H), 2.16 (p, <sup>3</sup>J<sub>HH</sub>=7.2 Hz, 2 H), 2.9-3.0 (m, 4 H), 3.68 (s, 1 H), 6.69 (s, 1 H), 7.3-7.5 (m, 4 H), 7.63 (d, <sup>3</sup>J<sub>HH</sub>=7.4 Hz, 2 H).

<sup>13</sup>C NMR (CDCl<sub>3</sub>): δ -0.32, -0.09, 26.28, 33.39, 34.11, 46.46, 47.54, 49.81, 115.80, 119.30, 126.92, 127.89, 128.46, 132.99, 137.30, 140.20, 140.81, 141.64, 142.08, 144.83.

6) Preparation of N-(1,1-Dimethylethyl)-1,1-dimethyl-1-(1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl) silanamine, dilithium salt.

N-(1,1-Dimethylethyl)-1,1-dimethyl-1-(1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl)silanamine (10.6551 g, 0.02947 moles) was stirred in hexane (100 mL) as nBuLi (0.070 moles, 35.00 mL of 2.0 M solution in cyclohexane) was added slowly. This mixture was then allowed to stir overnight during which time no salts crashed out of the dark red solution. After the reaction period the volatiles were removed and the residue quickly washed with hexane (2 x 50 mL). The dark red residue was then pumped dry and used without further purification or analysis (9.6517 g, 87.7percent yield).

7) Preparation of Dichloro[N-(1,1-dimethylethyl)-1,1-dimethyl-1-[(1,2,3,4,5-η)-1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl]silanaminato(2-)-N]titanium.

N-(1,1-Dimethylethyl)-1,1-dimethyl-1-(1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl)silanamine, dilithium salt (4.5355 g, 0.01214 moles) in THF (50 mL) was added dropwise to a slurry of TiCl<sub>3</sub>(THF)<sub>3</sub> (4.5005 g, 0.01214 moles) in THF (100 mL). This mixture was allowed to stir for 2 hours. PbCl<sub>2</sub> (1.7136 g, 0.006162 moles) was then added and the mixture allowed to stir for an additional hour. After the reaction period

the volatiles were removed and the residue extracted and filtered using toluene.

Removal of the toluene resulted in the isolation of a dark residue. This residue was then slurried in hexane and cooled to 0°C. The desired product was then isolated via filtration as a red-brown crystalline solid (2.5280 g, 43.5percent yield).

5 <sup>1</sup>H NMR (CDCl<sub>3</sub>): δ 0.71 (s, 3 H), 0.97 (s, 3 H), 1.37 (s, 9 H), 2.0-2.2 (m, 2 H), 2.9-3.2 (m, 4 H), 6.62 (s, 1 H), 7.35-7.45 (m, 1 H), 7.50 (t, <sup>3</sup>J<sub>HH</sub>=7.8 Hz, 2 H), 7.57 (s, 1 H), 7.70 (d, <sup>3</sup>J<sub>HH</sub>=7.1 Hz, 2 H), 7.78 (s, 1 H).

<sup>1</sup>H NMR (C<sub>6</sub>D<sub>6</sub>): δ 0.44 (s, 3 H), 0.68 (s, 3 H), 1.35 (s, 9 H), 1.6-1.9 (m, 2 H), 2.5-3.9 (m, 4 H), 6.65 (s, 1 H), 7.1-7.2 (m, 1 H), 7.24 (t, <sup>3</sup>J<sub>HH</sub>=7.1 Hz, 2 H), 7.61 (s, 1 H), 7.69 (s, 1 H), 7.77-7.8 (m, 2 H).

<sup>13</sup>C NMR (CDCl<sub>3</sub>): δ 1.29, 3.89, 26.47, 32.62, 32.84, 32.92, 63.16, 98.25, 118.70, 121.75, 125.62, 128.46, 128.55, 128.79, 129.01, 134.11, 134.53, 136.04, 146.15, 148.93.

<sup>13</sup>C NMR (C<sub>6</sub>D<sub>6</sub>): δ 0.90, 3.57, 26.46, 32.56, 32.78, 62.88, 98.14, 119.19, 121.97, 125.84, 127.15, 128.83, 129.03, 129.55, 134.57, 135.04, 136.41, 136.51, 147.24, 148.96.

8) Preparation of Dimethyl[N-(1,1-dimethylethyl)-1,1-dimethyl-1-[(1,2,3,4,5-η)-1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl]silanaminato(2-)-N]titanium.

20 Dichloro[N-(1,1-dimethylethyl)-1,1-dimethyl-1-[(1,2,3,4,5-η)-1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl]silanaminato(2-)-N]titanium (0.4970 g, 0.001039 moles) was stirred in diethyl ether (50 mL) as MeMgBr (0.0021 moles, 0.70 mL of 3.0 M solution in diethyl ether) was added slowly. This mixture was then stirred for 1 hour. After the reaction period the volatiles were removed and the residue extracted and filtered using hexane. Removal of the hexane resulted in the isolation of the  
25 desired product as a golden yellow solid (0.4546 g, 66.7 percent yield).

$^1\text{H}$  NMR ( $\text{C}_6\text{D}_6$ ):  $\delta$ 0.071 (s, 3 H), 0.49 (s, 3 H), 0.70 (s, 3 H), 0.73 (s, 3 H), 1.49 (s, 9 H), 1.7-1.8 (m, 2 H), 2.5-2.8 (m, 4 H), 6.41 (s, 1 H), 7.29 (t,  $^3J_{\text{HH}}=7.4$  Hz, 2 H), 7.48 (s, 1 H), 7.72 (d,  $^3J_{\text{HH}}=7.4$  Hz, 2 H), 7.92 (s, 1 H).

$^{13}\text{C}$  NMR ( $\text{C}_6\text{D}_6$ ):  $\delta$ 2.19, 4.61, 27.12, 32.86, 33.00, 34.73, 58.68, 58.82, 118.62, 121.98, 124.26, 127.32, 128.63, 128.98, 131.23, 134.39, 136.38, 143.19, 144.85.

#### Preparation of bis(hydrogenated-tallowalkyl)methylamine Cocatalyst

Methylcyclohexane (1200 mL) was placed in a 2L cylindrical flask. While stirring, 104 g, ground to a granular form of bis(hydrogenated-tallowalkyl)methylamine (ARMEEN<sup>®</sup> M2HT available from Akzo Chemical,) was added to the flask and stirred until completely dissolved. Aqueous HCl (1M, 200 mL) was added to the flask, and the mixture was stirred for 30 minutes. A white precipitate formed immediately. At the end of this time,  $\text{LiB}(\text{C}_6\text{F}_5)_4 \cdot \text{Et}_2\text{O} \cdot 3 \text{ LiCl}$  (Mw = 887.3; 177.4 g) was added to the flask. The solution began to turn milky white. The flask was equipped with a 6" Vigreux column topped with a distillation apparatus and the mixture was heated (140 °C external wall temperature). A mixture of ether and methylcyclohexane was distilled from the flask. The two-phase solution was now only slightly hazy. The mixture was allowed to cool to room temperature, and the contents were placed in a 4 L separatory funnel. The aqueous layer was removed and discarded, and the organic layer was washed twice with  $\text{H}_2\text{O}$  and the aqueous layers again discarded. The  $\text{H}_2\text{O}$  saturated methylcyclohexane solutions were measured to contain 0.48 wt percent diethyl ether ( $\text{Et}_2\text{O}$ ).

The solution (600 mL) was transferred into a 1 L flask, sparged thoroughly with nitrogen, and transferred into the drybox. The solution was passed through a column (1" diameter, 6" height) containing 13X molecular sieves. This reduced the level of  $\text{Et}_2\text{O}$  from 0.48 wt percent to 0.28 wt percent. The material was then stirred over fresh 13X sieves (20 g) for four hours. The  $\text{Et}_2\text{O}$  level was then measured to be 0.19 wt percent. The mixture was then stirred overnight, resulting in a further reduction in  $\text{Et}_2\text{O}$  level to approximately 40 ppm. The mixture was filtered using a funnel equipped with a glass frit having a pore size of 10-15  $\mu\text{m}$  to give a clear solution (the molecular sieves

were rinsed with additional dry methylcyclohexane). The concentration was measured by gravimetric analysis yielding a value of 16.7 wt percent.

### Polymerization

5           ESI #'s 7 - 31 were prepared in a 6 gallon (22.7 L), oil jacketed, Autoclave continuously stirred tank reactor (CSTR). A magnetically coupled agitator with Lightning A-320 impellers provided the mixing. The reactor ran liquid full at 475 psig (3,275 kPa). Process flow was in at the bottom and out of the top. A heat transfer oil was circulated through the jacket of the reactor to remove some of the heat of reaction.

10       At the exit of the reactor was a micromotion flow meter that measured flow and solution density. All lines on the exit of the reactor were traced with 50 psi (344.7 kPa) steam and insulated.

          Toluene solvent was supplied to the reactor at 30 psig (207 kPa). The feed to the reactor was measured by a Micro-Motion mass flow meter. A variable speed

15       diaphragm pump controlled the feed rate. At the discharge of the solvent pump, a side stream was taken to provide flush flows for the catalyst injection line (1 lb/hr (0.45 kg/hr)) and the reactor agitator (0.75 lb/hr (0.34 kg/hr)). These flows were measured by differential pressure flow meters and controlled by manual adjustment of micro-flow needle valves. Uninhibited styrene monomer was supplied to the reactor at 30 psig

20       (207 kPa). The feed to the reactor was measured by a Micro-Motion mass flow meter. A variable speed diaphragm pump controlled the feed rate. The styrene stream was mixed with the remaining solvent stream. Ethylene was supplied to the reactor at 600 psig (4,137 kPa). The ethylene stream was measured by a Micro-Motion mass flow meter just prior to the Research valve controlling flow. A Brooks flow meter/controller

25       was used to deliver hydrogen into the ethylene stream at the outlet of the ethylene control valve. The ethylene/hydrogen mixture combines with the solvent/styrene stream at ambient temperature. The temperature of the solvent/monomer as it enters the reactor was dropped to ~5 °C by an exchanger with -5°C glycol on the jacket. This stream entered the bottom of the reactor. The three component catalyst system and its

30       solvent flush also entered the reactor at the bottom but through a different port than the

monomer stream. Preparation of the catalyst components took place in an inert atmosphere glove box. The diluted components were put in nitrogen padded cylinders and charged to the catalyst run tanks in the process area. From these run tanks the catalyst was pressured up with piston pumps and the flow was measured with Micro-  
5 Motion mass flow meters. These streams combine with each other and the catalyst flush solvent just prior to entry through a single injection line into the reactor.

Polymerization was stopped with the addition of catalyst kill (water mixed with solvent) into the reactor product line after the micromotion flow meter measuring the solution density. Other polymer additives can be added with the catalyst kill. A static  
10 mixer in the line provided dispersion of the catalyst kill and additives in the reactor effluent stream. This stream next entered post reactor heaters that provide additional energy for the solvent removal flash. This flash occurred as the effluent exited the post reactor heater and the pressure was dropped from 475 psig (3,275 kPa) down to ~250mm of pressure absolute at the reactor pressure control valve. This flashed  
15 polymer entered a hot oil jacketed devolatilizer. Approximately 85 percent of the volatiles were removed from the polymer in the devolatilizer. The volatiles exited the top of the devolatilizer. The stream was condensed with a glycol jacketed exchanger and entered the suction of a vacuum pump and was discharged to a glycol jacket solvent and styrene/ethylene separation vessel. Solvent and styrene were removed from  
20 the bottom of the vessel and ethylene from the top. The ethylene stream was measured with a Micro-Motion mass flow meter and analyzed for composition. The measurement of vented ethylene plus a calculation of the dissolved gasses in the solvent/styrene stream were used to calculate the ethylene conversion. The polymer separated in the devolatilizer was pumped out with a gear pump to a ZSK-30  
25 devolatilizing vacuum extruder. The dry polymer exits the extruder as a single strand. This strand was cooled as it was pulled through a water bath. The excess water was blown from the strand with air and the strand was chopped into pellets with a strand chopper.

The various catalysts, co-catalysts and process conditions used to prepare the various individual ethylene styrene interpolymers (ESI #'s 7 - 31) are summarized in Table 3 and their properties are summarized in Table 4.

Table 3. Preparation Conditions for ESI #'s 7 - 31

ESI #	Reactor Temp °C	Solvent Flow lb/hr (kg/hr)	Ethylene Flow lb/hr (kg/hr)	Hydrogen Flow sccm	Styrene Flow lb/hr (kg/hr)	Ethylene Conversn %	B/Ti Ratio	MMAO <sup>e</sup> /Ti Ratio	Catalyst	Co-Catalyst
ESI 7	75.0	10.68 (4.84)	1.20 (0.54)	30.0	15.0 (6.8)	90.3	1.24	7.9	B <sup>b</sup>	C <sup>c</sup>
ESI 8	65.7	9.16 (4.15)	0.79 (0.36)	4.5	13.0 (5.9)	86.7	1.25	12.1	B <sup>b</sup>	C <sup>c</sup>
ESI 9	72.0	26.39 (11.97)	1.90 (0.86)	24.0	20.6 (9.3)	77.4	3.00	10.0	B <sup>b</sup>	D <sup>d</sup>
ESI 10	101.3	19.12 (8.67)	2.00 (0.91)	4.0	7.0 (3.2)	85.3	1.25	10.0	B <sup>b</sup>	D <sup>d</sup>
ESI 11	102.3	19.21 (8.71)	2.00 (0.91)	4.0	7.0 (3.2)	89.6	1.25	10.0	B <sup>b</sup>	C <sup>c</sup>
ESI 12	89.6	30.44 (13.80)	2.91 (1.32)	21.0	8.5 (3.9)	92.5	1.24	10.1	A <sup>a</sup>	C <sup>c</sup>
ESI 13	91.0	29.93 (13.57)	2.89 (1.31)	20.9	9.0 (4.1)	92.1	1.25	10.0	A <sup>a</sup>	C <sup>c</sup>
ESI 14	86.9	29.76 (13.50)	2.49 (1.13)	20.1	9.0 (4.1)	92.7	1.24	9.9	A <sup>a</sup>	C <sup>c</sup>
ESI 15	80.3	18.55 (8.41)	1.70 (0.77)	12.0	12.0 (5.4)	87.4	1.25	10.0	A <sup>a</sup>	C <sup>c</sup>
ESI 16	68.8	2.49 (1.13)	1.00 (0.45)	3.5	20.0 (9.1)	89.0	1.25	10.0	B <sup>b</sup>	C <sup>c</sup>
ESI 17	69.2	2.98 (1.35)	1.00 (0.45)	2.7	20.0 (9.1)	86.3	1.25	9.9	B <sup>b</sup>	C <sup>c</sup>
ESI 18	69.1	2.92 (1.32)	1.00 (0.45)	2.7	20.0 (9.1)	88.8	1.26	10.1	B <sup>b</sup>	C <sup>c</sup>
ESI 19	69.6	2.95 (1.34)	1.00 (0.45)	2.7	20.0 (9.1)	84.8	1.25	10.0	B <sup>b</sup>	C <sup>c</sup>
ESI 20	67.7	3.03 (1.37)	1.01 (0.46)	3.5	20.0 (9.1)	86.4	1.25	10.0	B <sup>b</sup>	C <sup>c</sup>
ESI 21	67.8	2.93 (1.33)	1.01 (0.46)	50.0	20.0 (9.1)	89.0	1.25	10.0	B <sup>b</sup>	C <sup>c</sup>
ESI 22	67.8	2.99 (1.36)	1.00 (0.45)	65.0	20.0 (9.1)	86.6	1.25	9.9	B <sup>b</sup>	C <sup>c</sup>
ESI 23	68.0	2.52 (1.14)	1.00 (0.45)	65.0	20.0 (9.1)	81.3	1.25	10.0	B <sup>b</sup>	C <sup>c</sup>

Table 3 Continued

ESI #	Reactor Temp °C	Solvent Flow lb/hr (kg/hr)	Ethylene Flow lb/hr (kg/hr)	Hydrogen Flow sccm	Styrene Flow lb/hr (kg/hr)	Ethylene Convers n. %	B/Ti Ratio	MMAO <sup>e</sup> /Ti Ratio	Catalyst	Co-Catalyst
ESI 24	69.1	5.89 (2.67)	1.01 (0.46)	15.0	15.0 (6.8)	87.9	1.25	8.1	B <sup>b</sup>	C <sup>c</sup>
ESI 25	67.1	2.43 (1.10)	1.20 (0.54)	0.0	23.8 (10.8)	90.85	1.24	10.0	B <sup>b</sup>	C <sup>c</sup>
ESI 26	98.7	50.00 (22.68)	4.35 (1.97)	24.8	5.0 (2.3)	96.5	3.50	3.5	A <sup>a</sup>	D <sup>d</sup>
ESI 27	93.6	38.01 (17.24)	3.10 (1.41)	13.2	6.9 (3.1)	96.3	3.00	7.0	A <sup>a</sup>	D <sup>d</sup>
ESI 28	78.8	31.56 (14.31)	1.74 (0.79)	4.0	13.5 (6.1)	95.3	3.50	9.0	A <sup>a</sup>	D <sup>d</sup>
ESI 29	77.5	41.00 (18.59)	2.18 (0.99)	30.0	16.5 (7.5)	94.1	3.5	9.0	A <sup>a</sup>	D <sup>d</sup>
ESI 30	75.1	41.00 (18.59)	2.17 (0.98)	3.8	21.0 (9.5)	97.6	3.5	6.0	A <sup>a</sup>	D <sup>d</sup>
ESI 31	72.1	15.93 (7.22)	1.20 (0.54)	31.9	10.0 (4.5)	90.3	1.2	8.0	A <sup>a</sup>	C <sup>c</sup>

a Catalyst A is dimethyl[N-(1,1-dimethylethyl)-1,1-dimethyl-1-[(1,2,3,4,5-η)-1,5,6,7-tetrahydro-3-phenyl-s-indacen-1-yl]silanaminato(2-)-N]-titanium.

5 b Catalyst B is (t-butylamido)dimethyl(tetramethylcyclopentadienyl)silane-titanium (II) 1,3-pentadiene prepared as described in U.S. Patent # 5,556,928, Example 17

c Cocatalyst C is bis-hydrogenated tallowalkyl methylammonium tetrakis (pentafluorophenyl)borate.

d Cocatalyst D is tris(pentafluorophenyl)borane, (CAS# 001109-15-5),.

e a modified methylaluminoxane commercially available from Akzo Nobel as MMAO-3A (CAS# 146905-79-5)



Table 4. Properties of ESI #'s 7 - 31

ESI #	ESI Styrene (wt%)	ESI Styrene (mol%)	Atactic Polystyrene (wt%)	Melt Index, I <sub>2</sub> (g/10m)	G # cm <sup>3</sup> /10m	10 <sup>3</sup> M <sub>w</sub>	M <sub>w</sub> /M <sub>n</sub> Ratio	T <sub>g</sub> (°C)
ESI 7	76	46	3.9	12.5	12.52	138	2.40	34.8
ESI 8	66	34	N/A	0.7	N/A	N/A	N/A	20.5
ESI 9	53	23	12.1	10.4	10.43	116	3.38	21.1
ESI 10	30	10	6	-	1.25	N/A	N/A	N/A
ESI 11	28	9	6.5	-	1.03	N/A	N/A	N/A
ESI 12	43.8	17.3	0.4	-	1.02	N/A	N/A	N/A
ESI 13	44.1	17.5	1.5	-	1.00	N/A	N/A	N/A
ESI 14	50	21	1.0	-	1.22	147	2.54	-10.0
ESI 15	58	27	3.3	-	0.98	236	2.37	-2.0
ESI 16	69	37	N/A	-	1.26	N/A	N/A	16.0
ESI 17	73	42	N/A	-	1.27	N/A	N/A	21.5
ESI 18	74	43	N/A	-	1.41	N/A	N/A	22
ESI 19	73.3	42	27.3	-	1.2	230	3.35	21.0
ESI 20	74.3	44	N/A	-	3.0	N/A	N/A	21.3
ESI 21	71.3	40	N/A	-	14.0	N/A	N/A	19.9
ESI 22	73.2	42	N/A	-	29.0	N/A	N/A	18.0
ESI 23	73.3	42	15.3	-	43.0	N/A	N/A	17.1
ESI 24	73.8	43	44.2	-	55.0	130	3.79	16.1
ESI 25	73.1	42	15.3	-	1.8	117	3.04	23.6
ESI 26	30.9	11	0.6	-	2.7	N/A	N/A	N/A
ESI 27	46.4	19	1.2	-	1.6	N/A	N/A	N/A
ESI 28	65.6	34	2.5	-	1.2	N/A	N/A	N/A
ESI 29	65.2	33	1.9	-	9.4	N/A	N/A	N/A
ESI 30	59.8	29	17.8	-	1.4	N/A	N/A	N/A
ESI 31	73	39	N/A	-	1.2	N/A	N/A	21.0

3) Preparation of ESI #'s 32-34

- ESI #'s 32 - 34 are substantially random ethylene/styrene interpolymers  
 5 prepared using the following catalyst and polymerization procedures.

Preparation of Catalyst B: (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)-  
 silanetitanium 1,4-diphenylbutadiene.

## 1) Preparation of lithium 1H-cyclopenta[1]phenanthrene-2-yl

To a 250 ml round bottom flask containing 1.42 g (0.00657 mole) of 1H-cyclopenta[1]phenanthrene and 120 ml of benzene was added dropwise, 4.2 ml of a 1.60 M solution of n-BuLi in mixed hexanes. The solution was allowed to stir overnight. The lithium salt was isolated by filtration, washing twice with 25 ml benzene and drying under vacuum. Isolated yield was 1.426 g (97.7 percent). <sup>1</sup>H NMR analysis indicated the predominant isomer was substituted at the 2 position.

## 2) Preparation of (1H-cyclopenta[1]phenanthrene-2-yl)dimethylchlorosilane

To a 500 ml round bottom flask containing 4.16 g (0.0322 mole) of dimethyldichlorosilane (Me<sub>2</sub>SiCl<sub>2</sub>) and 250 ml of tetrahydrofuran (THF) was added dropwise a solution of 1.45 g (0.0064 mole) of lithium 1H-cyclopenta[1]phenanthrene-2-yl in THF. The solution was stirred for approximately 16 hours, after which the solvent was removed under reduced pressure, leaving an oily solid which was extracted with toluene, filtered through diatomaceous earth filter aid (Celite™), washed twice with toluene and dried under reduced pressure. Isolated yield was 1.98 g (99.5 percent).

## 3. Preparation of (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamino)silane

To a 500 ml round bottom flask containing 1.98 g (0.0064 mole) of (1H-cyclopenta[1]phenanthrene-2-yl)dimethylchlorosilane and 250 ml of hexane was added 2.00 ml (0.0160 mole) of t-butylamine. The reaction mixture was allowed to stir for several days, then filtered using diatomaceous earth filter aid (Celite™), washed twice with hexane. The product was isolated by removing residual solvent under reduced pressure. The isolated yield was 1.98 g (88.9 percent).

## 4. Preparation of dilithio (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)silane

To a 250 ml round bottom flask containing 1.03 g (0.0030 mole) of (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamino)silane and 120 ml of benzene

was added dropwise 3.90 ml of a solution of 1.6 M n-BuLi in mixed hexanes. The reaction mixture was stirred for approximately 16 hours. The product was isolated by filtration, washed twice with benzene and dried under reduced pressure. Isolated yield was 1.08 g (100 percent).

5 5. Preparation of (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)silanetitanium dichloride

To a 250 ml round bottom flask containing 1.17 g (0.0030 mole) of  $\text{TiCl}_3 \cdot 3\text{THF}$  and about 120 ml of THF was added at a fast drip rate about 50 ml of a THF solution of 1.08 g of dilithio (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)silane. The mixture was stirred at about 20 °C for 1.5 h at which time 0.55 gm (0.002 mole) of solid  $\text{PbCl}_2$  was added. After stirring for an additional 1.5 h the THF was removed under vacuum and the residue was extracted with toluene, filtered and dried under reduced pressure to give an orange solid. Yield was 1.31 g (93.5 percent).

15 6. Preparation of (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)silanetitanium 1,4-diphenylbutadiene

To a slurry of (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)silanetitanium dichloride (3.48 g, 0.0075 mole) and 1.551 gm (0.0075 mole) of 1,4-diphenylbutadiene in about 80 ml of toluene at 70 °C was added 9.9 ml of a 1.6 M solution of n-BuLi (0.0150 mole). The solution immediately darkened. The temperature was increased to bring the mixture to reflux and the mixture was maintained at that temperature for 2 hrs. The mixture was cooled to about -20 °C and the volatiles were removed under reduced pressure. The residue was slurried in 60 ml of mixed hexanes at about 20 °C for approximately 16 hours. The mixture was cooled to about -25 °C for about 1 h. The solids were collected on a glass frit by vacuum filtration and dried under reduced pressure. The dried solid was placed in a glass fiber thimble and solid extracted continuously with hexanes using a soxhlet extractor. After 6 h a crystalline solid was observed in the boiling pot. The mixture was cooled to about

-20 °C, isolated by filtration from the cold mixture and dried under reduced pressure to give 1.62 g of a dark crystalline solid. The filtrate was discarded. The solids in the extractor were stirred and the extraction continued with an additional quantity of mixed hexanes to give an additional 0.46 gm of the desired product as a dark crystalline solid.

## 5 Polymerization

ESI #'s 32- 34 were prepared in a continuously operating loop reactor (36.8 gal). An Ingersoll-Dresser twin screw pump provided the mixing. The reactor ran liquid full at 475 psig (3,275 kPa) with a residence time of approximately 25 minutes. Raw materials and catalyst/cocatalyst flows were fed into the suction of the twin screw pump through injectors and Kenics static mixers. The twin screw pump discharged into a 2" diameter line which supplied two Chemineer-Kenics 10-68 Type BEM Multi-Tube heat exchangers in series. The tubes of these exchangers contained twisted tapes to increase heat transfer. Upon exiting the last exchanger, loop flow returned through the injectors and static mixers to the suction of the pump. Heat transfer oil was circulated through the exchangers' jacket to control the loop temperature probe located just prior to the first exchanger. The exit stream of the loop reactor was taken off between the two exchangers. The flow and solution density of the exit stream was measured by a MicroMotion.

Solvent feed to the reactor was supplied by two different sources. A fresh stream of toluene from an 8480-S-E Pulsafeeder diaphragm pump with rates measured by a MicroMotion flowmeter was used to provide flush flow for the reactor seals (20 lb/hr (9.1 kg/hr). Recycle solvent was mixed with uninhibited styrene monomer on the suction side of five 8480-5-E Pulsafeeder diaphragm pumps in parallel. These five Pulsafeeder pumps supplied solvent and styrene to the reactor at 650 psig (4,583 kPa). Fresh styrene flow was measured by a MicroMotion flowmeter, and total recycle solvent/styrene flow was measured by a separate MicroMotion flowmeter. Ethylene was supplied to the reactor at 687 psig (4,838 kPa). The ethylene stream was measured by a Micro-Motion mass flowmeter. A Brooks flowmeter/controller was used to deliver hydrogen into the ethylene stream at the outlet of the ethylene control valve. The ethylene/hydrogen mixture combined with the solvent/styrene stream at ambient

temperature. The temperature of the entire feed stream as it entered the reactor loop was lowered to 2°C by an exchanger with -10°C glycol on the jacket. Preparation of the three catalyst components took place in three separate tanks: fresh solvent and concentrated catalyst/cocatalyst premix were added and mixed into their respective run tanks and fed into the reactor via variable speed 680-S-AEN7 Pulsafeeder diaphragm pumps. As previously explained, the three component catalyst system entered the reactor loop through an injector and static mixer into the suction side of the twin screw pump. The raw material feed stream was also fed into the reactor loop through an injector and static mixer downstream of the catalyst injection point but upstream of the twin screw pump suction.

Polymerization was stopped with the addition of catalyst kill (water mixed with solvent) into the reactor product line after the Micro Motion flowmeter measuring the solution density. A static mixer in the line provided dispersion of the catalyst kill and additives in the reactor effluent stream. This stream next entered post reactor heaters that provided additional energy for the solvent removal flash. This flash occurred as the effluent exited the post reactor heater and the pressure was dropped from 475 psig (3,275 kPa) down to 450 mmHg (60 kPa) of absolute pressure at the reactor pressure control valve. This flashed polymer entered the first of two hot oil jacketed devolatilizers. The volatiles flashing from the first devolatilizer were condensed with a glycol jacketed exchanger, passed through the suction of a vacuum pump, and were discharged to the solvent and styrene/ethylene separation vessel. Solvent and styrene were removed from the bottom of this vessel as recycle solvent while ethylene exhausted from the top. The ethylene stream was measured with a MicroMotion mass flowmeter. The measurement of vented ethylene plus a calculation of the dissolved gases in the solvent/styrene stream were used to calculate the ethylene conversion. The polymer and remaining solvent separated in the devolatilizer was pumped with a gear pump to a second devolatilizer. The pressure in the second devolatilizer was operated at 5 mmHg (0.7 kPa) absolute pressure to flash the remaining solvent. This solvent was condensed in a glycol heat exchanger, pumped through another vacuum pump, and exported to a waste tank for disposal. The dry polymer (< 1000 ppm total volatiles)

was pumped with a gear pump to an underwater pelletizer with 6-hole die, pelletized, spin-dried, and collected in 1000 lb boxes.

The various catalysts, co-catalysts and process conditions used to prepare the various individual ethylene styrene interpolymers (ESI #'s 32 -34) are summarized in Table 5 and their properties are summarized in Table 6.

**Table 5. Preparation Conditions for ESI #'s 32 -34<sup>a</sup>**

ESI # 3826-	Reactor Temp °C	Solvent Flow lb/hr (kg/hr)	Ethylene Flow lb/hr (kg/hr)	Hydrogen Flow sccm	Styrene Flow lb/hr (kg/hr)	Ethylene Conversion %	Co- Catalyst	B/Ti Ratio	MMAO <sup>b</sup> /Ti Ratio
ESI 32	76.1	415 (188)	26 (12)	0	153 (69)	96	C <sup>c</sup>	5.3	10
ESI 33	76.0	415 (188)	26 (12)	0	152 (69)	96	C <sup>c</sup>	5.5	10
ESI 34	76.0	415 (188)	26 (12)	0	151 (68)	96	C <sup>c</sup>	5.5	10

<sup>a</sup> catalyst was (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)-silanetitanium 1,4-diphenylbutadiene.

<sup>b</sup> a modified methylaluminoxane commercially available from Akzo Nobel as MMAO-3A (CAS# 146905-79-5)

10 <sup>c</sup> cocatalyst C is tris(pentafluorophenyl)borane (CAS# 001109-15-5),.

**Table 6. Properties of ESI #'s 32-34**

ESI #	ESI Styrene (wt%)	ESI Styrene (mol%)	Atactic Polystyrene (wt%)	Melt Index, I <sub>2</sub> (g/10m)	10 <sup>-3</sup> M <sub>w</sub>	M <sub>w</sub> /M <sub>n</sub> Ratio	T <sub>g</sub> (°C)
ESI 32	77.6	48.3	7.8	4.34	153.3	2.7	31.80
ESI 33	77.7	48.4	7.8	4.17	165.7	2.7	31.65
ESI 34	77.7	48.4	7.8	4.13	168.2	2.9	31.51

#### Effect of Temperature on the Elastic Modulus of Substantially Random Interpolymers

15 The ESI samples were injection molded and their elastic modulus determined as function of temperature using an Instron tensile tester under ASTM Method D-638 at various temperatures. These data are summarized in Table 7.

Table 7. Elastic Modulus vs Temperature for ESI Samples

ESI (#)	Styrene (wt%)	Styrene (mol%)	I <sub>2</sub> (g/10min)	T <sub>g</sub> (°C)	Temp (°C)	10 <sup>-7</sup> G' Elastic Modulus (dynes/cm <sup>2</sup> )
ESI 1	73	42	1.8	24.7	1.8	959.0
					20.5	614.0
					31.1	15.7
					40.8	2.6
ESI 7	76	46	12.5	34.8	0.5	982.0
					20.0	28.0
					29.8	18.0
					39.8	3.3
ESI 8	66	34	0.7	20.5	0.4	817.0
					19.8	25.0
					29.8	2.2
					39.3	1.6
ESI 9	53	23	10.4	21.1	-18.5	684.0
					1.6	11.8
					21.6	0.5

These data demonstrate the rapid change in the modulus as the temperature is increased above the polymer T<sub>g</sub>.

5 Effect of Temperature on the Elongation of Substantially Random Interpolymers.

A sample of ESI 1 having a styrene content of 42 mol percent (73 wt percent) and a melt index (I<sub>2</sub>) of 1.8 g/10min was injection molded and its percent elongation determined as function of temperature using a using an Instron tensile tester under ASTM Method D-638. These data are summarized in Table 8.

Table 8. Elongation vs Temperature for ESI 1

ESI (#)	Styrene (wt%)	Styrene (mol%)	I <sub>2</sub> (g/10min)	Tg (°C)	Temp (°C)	Elongation (%)
ESI 1	73	42	1.8	24.7	23	220
					40	585

These data demonstrate the rapid increase in percent elongation as the temperature is increased above the polymer Tg .

5

Effect of Styrene Content on the Tg of Substantially Random Ethylene/Styrene Interpolymers

The Tg of a series of substantially random ethylene/styrene interpolymers having similar molecular weight (G # ~ 1.0) was measured and the data are summarized in Table 9.

10

Table 9. Tg vs Styrene Content of Substantially Random Ethylene/Styrene Interpolymers

ESI #	Styrene (wt%)	Styrene (mol%)	Tg (°C)
ESI 15	58	27	-2
ESI 16	69	37	16
ESI 17	73	42	21
ESI 18	74	43	22
ESI 10/11*	27	9	-18
ESI 12/13*	40	15	-16
ESI 14	50	21	-10

\* 50:50 wt percent blend

15

The data in Table 9 demonstrate the increase in the polymer Tg as the styrene content of the substantially random ethylene/styrene interpolymers increases.



Effect of Molecular Weight on the Tg of Substantially Random Ethylene/Styrene Interpolymers

The Tg of a series of substantially random ethylene/styrene interpolymers having similar styrene content and a molecular weight as measured by Gottfert melt index, was determined and the data are shown in Table 10.

Table 10. Tg vs Gottfert # of Substantially Random Ethylene/Styrene Interpolymers

ESI #	Styrene (wt%)	Styrene (mol%)	Gottfert I <sub>2</sub> (cm <sup>3</sup> /10min)	Tg (°C)
ESI 19	73.3	42	1.2	21.0
ESI 20	74.3	44	3.0	21.3
ESI 21	71.3	40	14.0	19.9
ESI 22	73.2	42	29.0	18.0
ESI 23	73.3	42	43.0	17.1
ESI 24	73.8	43	55.0	16.1

The data in Table 10 demonstrate the increase in the polymer Tg as the molecular weight of the substantially random ethylene/styrene interpolymers increases.

Effect of Added Tackifiers on the Tg and Modulus of Substantially Random Ethylene/Styrene Interpolymers

The tackifiers evaluated in the study, as well as properties obtained from trade literature, are set forth in the following Table 11.

Table 11. Summary of Properties of Tackifiers Used in Present Invention

Tackifier	Manufacturer	Feedstock	Mn	Tg (°C)
Endex 155	Hercules	Copolymer Modified Styrene	2,900	100
Piccotex 120	Hercules	Copolymer Modified Styrene	1,600	68
Regalrez 1139	Hercules	Hydrogenated Styrenic	1,500	80
Kristalex 5140	Hercules	Copolymer of pure monomer	1450	88
Plastolyn 140	Hercules	Hydrogenated aliphatic hydrocarbon	370	90

A series of blends of ESI and various tackifiers were prepared in a Haake torque rheometer and the Tg of the various blends was measured. These data are summarized in Table 12.

Table 12. Effect of 10 wt percent of Various Tackifiers on Tg of ESI # 25 (42 mol percent styrene, 1.8 g/cm<sup>3</sup> Gottfert, Tg = 23.6°C)

Tackifier	Tg of Tackifier (°C)	Tg of Blend (°C)
Regalrez™ 1139	80.0	23.4
Picotex™ 120	68.0	25.0
Kristalex™ 5140	88.0	25.2
Plastolyn™ 140	90.0	25.6
Endex™ 155	100.0	25.7

The data in Table 12 demonstrate that the Tg of the substantially random ethylene/styrene interpolymers increases with the addition of the tackifiers used in the present invention.

The modulus of ESI 25 and a blend of ESI 25 and Endex 155 tackifier was measured as a function of temperature and the results are summarized in Table 13.

Table 13. Effect of 10 wt percent of Endex 155 on Modulus of  
ESI # 25 (42 mol percent styrene, 1.8 Gottfert, T<sub>g</sub> = 23.6°C)

Tackifier	Temperature (°C)	Modulus (Psig)
None	20.0	11,600
	33.0	290
Endex <sup>TM</sup> 155	20.0	4300
	33.0	290

The data in Table 14 demonstrate that the modulus of the substantially random ethylene/styrene interpolymers decreases with the addition of the tackifiers used in the present invention.

#### Examples 1-5

Fibers were produced by extruding the interpolymer using a one inch diameter extruder which feeds a gear pump. The gear pump pushes the material through a spin pack containing a 40 micrometer (average pore size) sintered flat metal filter and a 34 or 108 hole spinneret. The spinneret holes have a diameter of 400 or 800 micrometers both having a land length (i.e, length/diameter or L/D) of 4/1. The gear pump is operated such that about 0.39 grams of polymer are extruded through each hole of the spinneret per minute. The melt temperature of the polymer is typically from about 200 - 240°C, and varies depending upon the molecular weight and styrene content of the interpolymer being spun. Generally the higher the molecular weight, the higher the melt temperature. Quench air (about 25°C) is used to help the melt spun fibers cool. The quench air is located just below the spinneret and blows air across the fiber line as it is extruded. The quench air flow rate is low enough so that it can barely be felt by hand in the fiber area below the spinneret. The fibers are collected on a godet roll located about 3 meters below the spinneret die and having a diameter of about 6 inches (15.24 cm). The godet roll speed is adjustable, but for the experiments demonstrated herein, the godet speed ranged from about 200 - 3100 revolutions/minute.

Fibers were tested on an Instron tensile testing device equipped with a small plastic jaw on the cross-head (the jaw has a weight of about six gms) and a 500 gram load cell. The jaws are set 1 inch (2.54 cm) apart. The cross head speed is set at 5

inches/minute (12.7 cm/minute). A single fiber is loaded into the Instron jaws for testing. The fiber is then stretched to 100 percent of strain (that is, it is stretched another 1 inch), where the tenacity is recorded. The fiber is allowed to return to the original Instron setting (where the jaws are again 1 inch apart) and the fiber is again  
 5 pulled. At the point where the fiber begins to provide stress resistance, the strain is recorded and the percent permanent set is calculated.

Thus, a fiber pulled for the second time which did not provide stress resistance (that is, pull a load) until it had traveled 0.1 inches (0.25 cm) would have a percent permanent set is of 10 percent, that is, the percent of strain at which the fiber begins to  
 10 provide stress resistance. The numerical difference between the percent permanent set and 100 percent is known as the percent elastic recovery. Thus, a fiber having a permanent set of 10 percent will have a 90 percent elastic recovery. After recording percent permanent set, the fiber is pulled to 100 percent strain and the tenacity recorded. The fiber pulling process is repeated several times, with the percent  
 15 permanent set recorded each time and the 100 percent strain tenacity recorded as well. Finally, the fiber is pulled to its breaking point and the ultimate breaking tenacity and elongation recorded.

Table 14. Fiber Data for Examples 1 -5

Example #	ESI #	400um die 200°C drawdown (RPM)	400um die 220°C drawdown (RPM)	400um die 240°C drawdown (RPM)	800um die 200°C drawdown (RPM)	800um die 220°C drawdown (RPM)	800um die 240°C drawdown (RPM)
Ex. 1	26	not draw	300	not draw	200	200	400
Ex. 2	27	not draw	>200	800	>250	>250	800
Ex. 3	28	not draw	>250	1800	>250	300	400
Ex. 4	29	3100	3100	not draw	3100	3100	3000
Ex. 5	30	N/A	N/A	-1400	N/A	N/A	1600

20

#### Example 6

A sample of ESI 7 was spun on a laboratory fiber line using standard conditions. ESI 7 contained 46 mol percent styrene (76.0 wt percent) and had a Gottfert melt

index # (ml/10 min) of 12.5 and a Tg as measured by DSC of 34.8°C. The fibers from ESI 7 were flexed and were found to be stiff at the temperature of the lab (20°C).

#### Example 7

- 5           A sample of ESI 19 was spun on a laboratory fiber line as for Example 1. ESI 19 contained 73.3 weight percent styrene (42 mol percent) and had a Gottfert melt index # (ml/10 min) of 1.2 and a Tg as measured by DSC of 21.0°C.

#### Example 8

- 10           A sample of ESI 24 was spun on a laboratory fiber line as for Example 1. ESI 24 contained 73.8 weight percent styrene (73.3 mol percent) and had a Gottfert melt index # (ml/10 min) of 55.0 and a Tg as measured by DSC of 16.1°C.

#### Example 9

- 15           A sample of ESI 22 was spun on a laboratory fiber line as for Example 1. ESI 22 contained 73.2 weight percent styrene (42 mol percent) and had a Gottfert melt index # (ml/10 min) of 29.0 and a Tg as measured by DSC of 18.0°C.

Table 15. Fiber Data for Examples 6 - 9.

Example #	ESI #	Styrene (wt%)	Styrene (mol%)	Gottfert I2 (cm <sup>3</sup> /10min)	Tg (°C)
6	ESI 7	76.0	46	12.5	34.8
7	ESI 19	73.3	42	1.2	21.0
8	ESI 24	73.8	43	55.0	16.1
9	ESI 22	73.2	42	29.0	18.0

20

#### Examples 10 - 16

Fibers were prepared using ethylene/styrene interpolymers prepared essentially as for ESI's 7 -31 having the G #'s and styrene contents summarized in Table 16.

Examples 10 to 14 were tumble blended (dry blended) prior to fiber conversion. Examples 15 and 16 were prepared as melt blended blends in a Haake torque rheometer. The fibers were produced from these formulations under the following conditions:

5                                      Temperature set points:        160°C/230°C/250°C/250°C/250°C  
    Gear Pump Settings:            10 rpm and 2lb/hr throughput  
    Quench:                            Off  
    Haul Off                            700 rpm at 1.5-2.0 mil

10                                      The presence of additives in the formulations caused the haul off maximum speeds to decrease by at least 300 rpm. In other words to make a sample containing additives at 700 rpm haul off would require that the base resin be able to sustain a 1000 rpm haul off rate.

   The Tg values for the formulations are also summarized in Table 16.

15                                      Table 16. Results of Fiber Tests for Examples 10 - 16

Example #	Styrene (wt%)	Styrene (mol%)	aPS (wt%)	G# (ml/10min)	Additives (wt%)	Tg, (DSC) (°C)
Ex. 10	74.2	43.6	5	9.0	None	23.68
Ex. 11	74.2	43.6	5	9.0	Acrylic (10%) Tackifier* (20%)	27.47
Ex. 12	74.2	43.6	5	9.0	Tackifier* (30%)	34.71
Ex. 13	74.2	43.6	5	9.0	Acrylic (10%) Tackifier* (30%)	35.40
Ex. 14	75.0	44.7		2.3	None	28.67
Ex. 15	73.8	43.1		2.3	Acrylic (10%) Tackifier* (10%)	30.93
Ex. 16	73.2	42.4		2.3	Acrylic (10%) Tackifier* (20%)	32.60

\* Endel™ 155 tackifier

These results demonstrate that the Tg increases with added tackifier in the presence of 10 wtpercent acrylic

Effect of Added Tackifiers and a Second Blend Component on the Tg of Substantially Random Ethylene/Styrene Interpolymers

Examples 17 - 21

- 5        Examples 17 - 21 are fibers prepared as for Example 1 from a blend of ESI 25 having a styrene content of 42 mol percent (73.1 wt percent) and a Gottfert melt index of 1.8 g/cm<sup>3</sup> with Endex TM 155 tackifier and/or acrylic in the relative proportions summarized in Table 17. The blends were prepared as for Examples 10-14.

- 10        Table 17. Effect of Endex<sup>TM</sup> 155 and Acrylic on the Tg of Fibers Prepared from Blends With ESI # 25 (42 mol percent styrene, 1.8 g/cm<sup>3</sup> Gottfert, Tg = 23.6°C)

Example #	ESI #25 (wt%)	Acrylic (wt%)	Endex <sup>TM</sup> 155 (wt%)	Tg of Blend (°C)
Ex. 17	100	0	0	23.6
Ex. 18	90	10	0	22.7
Ex. 19	90	0	10	25.0
Ex. 20	80	10	10	24.2
Ex. 21	70	10	20	28.1

- 15        The data in Table 14 demonstrate that the Tg of the substantially random ethylene/styrene interpolymers increases with the addition of the tackifier and the second polymer component described and used in the present invention.

Effect of Endex<sup>TM</sup> 155 and Acrylic on Modulus at 20°C C and 33°C of ESI 25

Examples 22 - 25

- 20        A series of fibers were prepared as for Example 1 from blends of ESI 25, Endex<sup>TM</sup> 155 and Acrylic (PMMA) and the modulus measured at 20°C and 33°C. The blend compositions and modulus data are summarized in Table 18.

- 25        Table 18. Effect of Endex<sup>TM</sup> 155 and Acrylic on Modulus at 20°C and 33°C of Fibers Made From ESI 25 (73 wt percent styrene, 1.8 g/cm<sup>3</sup> Gottfert, Tg = 23.6°C)

Example #	ESI # 25 (wt%)	Endex™ 155 (wt%)	Acrylic (wt%)	Modulus at 20°C (psi)	Modulus at 33°C (psi)
Example 22	100	0	0	87,000	
Example 23	70	20	10	140,000	
Example 24	100	0	0		2,900
Example 25	70	20	10		58,000

The data in Table 18 demonstrate that both the ESI interpolpolymer and its blend with 10 wt % acrylic and 20 wt percent Endex™ 155 have an equivalent change in modulus above and below the T<sub>g</sub>.

5 Examples 26 - 28

A series of fibers were prepared as for Example 1 from ESI #'s 32 - 34. The T<sub>g</sub> data are summarized in Table 19.



Table 19. Fiber Data for Examples 26 - 28

Example #	ESI #	Styrene (wt%)	Styrene (mol%)	Gottfert I <sub>2</sub> (cm <sup>3</sup> /10min)	I <sub>2</sub> (g/10min)	Tg (°C)
26	ESI 32	77.1	47.5	3.48	4.34	31.8
27	ESI 33	76.4	46.6	3.35	4.17	31.7
28	ESI 34	84.4	59.3	3.31	4.13	31.5

These data show the increase in Tg observed for samples prepared from these interpolymers.

5 Examples 29-43

A series of bicomponent fibers were prepared from ESI 35 and the following second polymer components:

PP1 - a 35 MFR Polypropylene available from Montell having the product designation PF 635

- 10 PET1 - a Polyester available from Wellman having the product designation Blend 9869, lot# 61418.

PE1 - a linear low density ethylene/octene copolymer having a melt index, I<sub>2</sub>, of 17.0 g/10 min and a density of 0.950 g/cm<sup>3</sup>.

- 15 SAN2 - a styrene-acrylonitrile copolymer available from Dow Chemical having the product designation TYRIL™ 100.

The substantially random ethylene/styrene copolymer ESI 35 was prepared using the same catalyst and polymerization procedures as ESI's 32-34 using the process conditions in Table 20. ESI 35 had a melt index, I<sub>2</sub> of 0.94 g/10 min, an interpolymers  
 20 styrene content of 77.42 wt percent ( 48.0 mol percent) and an atactic polystyrene content of 7.48 wt percent, and contained 0.24 wt percent talc and 0.20 wt percent siloxane binder.

Table 20

ESI #	Reactor Temp °C	Solvent Flow lb/hr (kg/hr)	Ethylene Flow lb/hr (kg/hr)	Hydrogen Flow sccm	Styrene Flow lb/hr (kg/hr)	Ethylene Conversion %	B/Ti Ratio	MMAO/ Ti Ratio
ESI 35	57	755 (342)	33 (15)	100	243 (110)	98	4	8

A series of sheath core bicomponent fibers were produced by coextruding a substantially random ethylene/styrene interpolymer (ESI-35) as the core and a second polymer as the sheath. The fibers were fabricated using two 1.25 inch diameter extruders which fed two gear pumps each pumping at a rate of 6 cm<sup>3</sup>/rev multiplied by the meter pump speed in rpm (given in Table 21). The gear pumps pushed the material through a spin pack containing a filter and a multiple hole spinneret. The spin head temperature was typically from about 275 - 300°C, and varied depending upon the melting point and degradation temperature of the polymer components being spun. Generally the higher the molecular weight of the polymers, the higher the melt temperature. Quench air (about 10 to about 30°C) was used to help the melt spun fibers cool. The quench air was located just below the spinneret and blows air perpendicularly across the length of the fibers as they are extruded. The fibers were collected on a series of godet rolls to produce the yarn. The first godet located about 2.5 meters below the spinneret die and having a diameter of about 6 inches (15.24 cm). The godet roll speeds were adjustable, but for Examples 29 - 43, the godet speeds ranged from 100 to 1000 meters/minute. The compositions and fabrication conditions for the fibers of Examples 29 - 43 are summarized in Table 21. All examples are round core sheath bicomponent fibers with the exception of Example 39 which had a delta core sheath configuration.

Table 21

	Ex. 29	Ex. 30	Ex. 31	Ex. 32	Ex. 33	Ex. 34	Ex. 35	
Bico Configuration	Core	Core	Core	Core	Core	Core	Core	Sheath
Polymer Type	ESI	ESI	ESI	ESI	PP	PP	ESI	PE
Polymer Ratio (wt. %)	50	50	70	70	30	30	50	50
Extruder Temp. Zone 1 (°F)	220	217	217	217	200	200	220	200
Extruder Temp. Zone 2 (°F)	227	227	227	227	210	210	220	210
Extruder Temp. Zone 3 (°F)	270	270	270	270	220	220	270	225
Extruder Temp. Zone 4 (°F)	275	275	275	275	240	240	275	240
Melt Temperature (°F)	282	286	286	286	286	286	284	282
Extruder Pressure (psi)	750	750	750	750	750	750	750	750
Pack Pressure (psi)	2070	2720	2720	2860	1210	1290	2270	1570
Meter Pump Speed (rpm)	5.13	8.22	8.91	12.18	5.1	6.97	4.33	4.56
Extruder amps (A)	4.2	4	5.3	6.1	3	3.1	4.5	2.7
Denier Roll Speed (mpm)	151	151	151	151			125	
Tension Roll Speed (mpm)	151	152	152	152			127	
Draw Roll #1 Speed / Temp (mpm / °C)	151/50	152/50	152/65	152/65			128/65	
Draw Roll #2 Speed / Temp (mpm / °C)	306/50	551/50	551/65	551/65			350/65	
Relax Roll Speed / Temp (mpm / °C)	292/25	534/25	534/25	534/25			250/65	
Spin Head Temperature (°C)	295	295	295	295			295	
Quench Air Temperature (°F)	68	68	68	68			68	

Table 21 Continued

	Ex. 36	Ex. 37	E. 38	Ex. 39	Ex. 40	Ex. 41	Ex. 42	Ex. 43	
Bico Configuration	Core	Sheat	Core	Δ Core	Δ Sheath	Core	Sheath	Core	Sheath
Polymer Type	ESI	PE	ESI	PE	ESI	PET	PET	ESI	PET
Polymer Ratio (wt. %)	60	40	70	30	70	30	10	90	10
Extruder Temp. Zone 1 (°F)	220	200	220	200	216	289	290	220	230
Extruder Temp. Zone 2 (°F)	220	210	220	210	222	290	295	220	235
Extruder Temp. Zone 3 (°F)	270	225	270	225	269	291	291	270	240
Extruder Temp. Zone 4 (°F)	275	240	275	240	275	294	294	275	240
Melt Temperature (°F)	284	282	284	282	297	295	298	270	268
Extruder Pressure (psi)	750	750	750	750	750	750	750	750	750
Pack Pressure (psi)	2430	1520	2610	2640	2130	1037	990	2200	450
Meter Pump Speed (rpm)	5.2	36.5	6.06	14.56	14.36	5.66	2.1	14.4	1.6
Extruder amps (A)	4.1	2.5	4.6	4	4.7	3	2.5	4.7	12.5
Denier Roll Speed (mpm)	125	125	125	125	250			200	
Tension Roll Speed (mpm)	127	127	127	127	251			203	
Draw Roll #1 Speed / Temp (mpm / °C)	128/65	128/65	128/6	129/65	252/65			203/65	
Draw Roll #2 Speed / Temp (mpm / °C)	260/65	290/65	260/6	390/65	807/65			606/65	
Relax Roll Speed / Temp (mpm / °C)	250/25	250/25	250/25	350/25	794/25			604/25	
Spin Head Temperature (°C)	295	295	295	295	300			300	
Quench Air Temperature (°F)	68	68	68		66			52	

Approximately 45 m of the resulting yarn was transferred to a denier wheel which was then weighed to determine the number of denier per filament. The resulting yarn were tested on an Model 100 INSTRON tensile testing device equipped with a type 4C (INSTRON #2714-004, 150 lb cap./90 psi max) jaw on the cross-head and a 100 lb load cell. The cross head speed was set at 130 mm/min. The yarn was loaded into the Instron jaws for testing. The yarn was then stretched to break and the ultimate breaking tenacity and elongation were recorded. The results of the testing are summarized in Table 22.

Table 22.

Bicomponent Fiber Properties<sup>+</sup>

Example #	Denier (dn)	Tenacity (g/dn)	Elongation (%)
29	1127 (1130)	1.12 (1.10)	146 (140)
30	1186 (1190)	1.81 (1.80)	56 (50)
31	950 (952)	2.10 (1.90)	92 (88)
32	1230 (1238)	1. (1.60)	86 (80)
33	826 (823)	1.13 (1.30)	121 (103)
34	1256 (1261)	0.87 (0.83)	162 (186)
35	1227 (1226)	0.80 (0.67)	217 (207)
36	1224 (1222)	0.93 (1.05)	130 (140)
37	840 (874)	1.50 (1.10)	186 (127)
38	1224 (1217)	0.96 (0.92)	200 (184)
39	1110 (1083)	1.33 (1.13)	144 (150)
40	1170 (1174)	2.31 (2.30)	71 (69)
41	954 (534)	1.16 (1.80)	61 (53)
42	1460 (1450)	1.55 (1.25)	151 (85)
43	*	*	*

+ values in parentheses represent same measurements made after 48 hr.

\* the data generated from this example had too much variability to accurately determine a value.

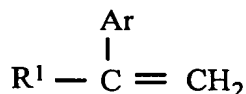
These results demonstrate that bicomponent fibers can be prepared with improved tenacity ( $\geq 0.8$  g/dn) which remains, along with other physical properties, relatively unchanged over time. Thus choice of the sheath component can be used to

instill the physical properties of the fiber while the choice core component can be used to exert an influence on the elongation and other stress strain characteristics.

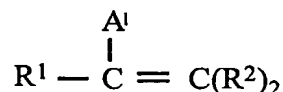
## CLAIMS:

- 1 1. A fiber comprising;  
 2 (A) from 50 to 100 wt percent (based on the combined weights of Components  
 3 A and B) of at least one substantially random interpolmer having an  $I_2$  of  
 4 from 0.1 to 1,000 g/10 min, a density greater than 0.9300 g/cm<sup>3</sup>, and an  
 5  $M_w/M_n$  of 1.5 to 20; which comprises;  
 6 (1) from 0.5 to 65 mol percent of polymer units derived from;  
 7 (i) at least one vinyl or vinylidene aromatic monomer, or  
 8 (ii) at least one hindered aliphatic or cycloaliphatic vinyl or  
 9 vinylidene monomer, or  
 10 (iii) a combination of at least one aromatic vinyl or vinylidene  
 11 monomer and at least one hindered aliphatic or cycloaliphatic  
 12 vinyl or vinylidene monomer, and  
 13 (2) from 35 to 99.5 mol percent of polymer units derived from ethylene  
 14 or at least one C<sub>3-20</sub>  $\alpha$ -olefin, or a combination thereof; and  
 15 (B) from 0 to 50 percent by weight (based on the combined weights of  
 16 Components A and B) of at least one tackifier.

- 1 2. The fiber of Claim 1 wherein;  
 2 (A) Component (A) is present in an amount of 50 to 95 wt percent (based on  
 3 the combined weights of Components A and B) and comprises at least one  
 4 substantially random interpolmer having an  $I_2$  of 0.5 to 200 g/10 min, a  
 5 density of from 0.930 to 1.045 g/cm<sup>3</sup> and an  $M_w/M_n$  of 1.8 to 10; which  
 6 comprises;  
 7 (1) from 1 to 55 mol percent of polymer units derived from;  
 8 (i) said vinyl or vinylidene aromatic monomer represented by  
 9 the following formula;



wherein R<sup>1</sup> is selected from the group of radicals consisting of hydrogen and alkyl radicals containing three carbons or less, and Ar is a phenyl group or a phenyl group substituted with from 1 to 5 substituents selected from the group consisting of halo, C<sub>1-4</sub>-alkyl, and C<sub>1-4</sub>-haloalkyl; or  
 (ii) said hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer represented by the following general formula;



wherein A<sup>1</sup> is a sterically bulky, aliphatic or cycloaliphatic substituent of up to 20 carbons, R<sup>1</sup> is selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; each R<sup>2</sup> is independently selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; or alternatively R<sup>1</sup> and A<sup>1</sup> together form a ring system; and  
 (2) from 45 to 99 mol percent of polymer units derived from ethylene, or ethylene and at least one of propylene, 4-methyl-1-pentene, butene-1, hexene-1 or octene-1; and

B) said tackifier, Component B, is present in an amount from 5 to 50 percent by weight (based on the combined weights of components A and B) and comprises a wood rosin, a tall oil derivative, a cyclopentadiene derivative, a natural terpene, a synthetic terpene, a terpene-phenolic resin, a styrene/α-methyl styrene resin, or a mixed aliphatic-aromatic tackifying resin, or any combination thereof.

3. The fiber of Claim 1 wherein;

(A) Component (A) is present in an amount of 60 to 90 wt percent (based on the combined weights of Components A and B) and comprises at least one



4 substantially random interpolymer having an  $I_2$  of 0.5 to 100 g/10 min, a  
5 density of from 0.930 to 1.040 g/cm<sup>3</sup> and an  $M_w/M_n$  of 2 to 5; which  
6 comprises;

7 (1) from 2 to 50 mol percent of polymer units derived from;

8 i) said vinyl or vinylidene aromatic monomer which comprises  
9 styrene,  $\alpha$ -methyl styrene, ortho-, meta-, and para-  
10 methylstyrene, and the ring halogenated styrenes, or

11 ii) said hindered aliphatic or cycloaliphatic vinyl or vinylidene  
12 monomers which comprises 5-ethylidene-2-norbornene or 1-  
13 vinylcyclo-hexene, 3-vinylcyclo-hexene, and 4-  
14 vinylcyclohexene;

15 (2) from 50 to 98 mol percent of polymer units derived from ethylene,  
16 or ethylene and at least one of propylene, 4-methyl-1-pentene,  
17 butene-1, hexene-1 or octene-1; and

18 B) said tackifier, Component B, is present in an amount from 10 to 40 percent  
19 by weight (based on the combined weights of components A and B) and  
20 comprises a styrene/ $\alpha$ -methyl styrene resin, or a mixed aliphatic-aromatic  
21 tackifying resin or any combination thereof.

1 4. The fiber of Claim 3 wherein Component A1 is styrene; and Component A2 is  
2 ethylene and at least one of propylene, 4-methyl-1-pentene, butene-1, hexene-1 or  
3 octene-1; and Component B is a styrene/ $\alpha$ -methyl styrene resin.

1 5. The fiber of Claim 3 wherein Component A1 is styrene, Component A2 is ethylene,  
2 Component B is a styrene/ $\alpha$ -methyl styrene resin.

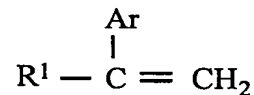
1 6. The fiber of Claim 1, in blended form with other forms of fibers.

1 7. The fiber of Claim 6, blended with cotton fibers.

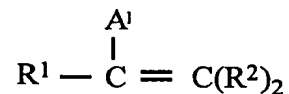
- 1 8. The fiber of Claim 7, blended with polyester fibers.
- 1 9. A fabric comprising the fiber of Claim 1.
- 1 10. The fabric of Claim 9, comprising a woven fabric.
- 1 11. The fabric of Claim 9, comprising a non-woven fabric.
- 1 12. A fabricated article prepared from the fiber of Claim 1, comprising carpet, doll  
2 hair, a tampon, a diaper, athletic sportswear, wrinkle free and form-fitting apparel,  
3 upholstery, bandages, and gamma sterilizable non-woven articles.
- 1 13. A plurality of the fibers of Claim 1 in the form of doll hair.
- 1 14. A bicomponent fiber comprising;  
2 (I) a first component comprising from 5 to 95 wt percent (based on the  
3 combined weights of Components I and II) of  
4 (A) from 50 to 100 wt percent (based on the combined weights of  
5 Components A and B) of at least one substantially random  
6 interpolymer having an  $I_2$  of from 0.1 to 1,000 g/10 min, a density  
7 greater than 0.9300 g/cm<sup>3</sup>, and an  $M_w/M_n$  of 1.5 to 20; which  
8 comprises;  
9 (1) from 0.5 to 65 mol percent of polymer units derived from;  
10 (a) at least one vinyl or vinylidene aromatic monomer, or  
11 (b) at least one hindered aliphatic or cycloaliphatic vinyl or  
12 vinylidene monomer, or  
13 (c) a combination of at least one aromatic vinyl or vinylidene  
14 monomer and at least one hindered aliphatic or cycloaliphatic  
15 vinyl or vinylidene monomer, and

- 16 (2) from 35 to 99.5 mol percent of polymer units derived from ethylene  
17 or at least one C<sub>3-20</sub> α-olefin or a combination thereof; and  
18 (B) from 0 to 50 percent by weight (based on the combined weights of  
19 Components A and B) of at least one tackifier; and  
20 (II) a second component, present in amount of from 5 to 95 wt percent  
21 (based on the combined weights of Components I and II) which  
22 comprises one or more of;  
23 A) an ethylene or α-olefin homopolymer or interpolymers;  
24 B) an ethylene/propylene rubber (EPM), ethylene/propylene  
25 diene monomer terpolymer (EPDM) , isotactic  
26 polypropylene;  
27 C) a styrene/ethylene-butene copolymer, a styrene/ethylene-  
28 propylene copolymer, a styrene/ethylene-butene/styrene  
29 (SEBS) copolymer, a styrene/ethylene-propylene/styrene  
30 (SEPS) copolymer,  
31 D) the acrylonitrile-butadiene-styrene (ABS) polymers,  
32 styrene-acrylonitrile (SAN), high impact polystyrene,  
33 E) polyisoprene, polybutadiene, natural rubbers,  
34 ethylene/propylene rubbers, ethylene/propylene diene  
35 (EPDM) rubbers, styrene/butadiene rubbers, thermoplastic  
36 polyurethanes,  
37 F) epoxies, vinyl ester resins, polyurethanes, phenolic resins,  
38 G) homopolymers or copolymers of vinyl chloride or  
39 vinylidene chloride,  
40 H) poly(methylmethacrylate), polyester, nylon-6, nylon-6,6,  
41 poly(acetal); poly(amide), poly(arylate), poly(carbonate),  
42 poly(butylene) and polybutylene, polyethylene  
43 terephthalates.

- 1 15. The bicomponent fiber of Claim 14 which is of the sheath/core type, segmented  
 2 pie type, side-by-side or "islands in the sea" type; and wherein;
- 3 (i) said first component I comprises from 25 to 95 wt percent (based on the  
 4 combined weights of Components I and II);
- 5 (ii) Component I(A) is present in an amount of 50 to 95 wt percent (based on  
 6 the combined weights of Components IA and IB) and comprises at least  
 7 one substantially random interpolymer having an  $I_2$  of 0.5 to 200 g/10 min,  
 8 a density of from 0.930 to 1.045 g/cm<sup>3</sup> and an  $M_w/M_n$  of 1.8 to 10; which  
 9 comprises;
- 10 (1) from 1 to 55 mol percent of polymer units derived from;
- 11 (a) said vinyl or vinylidene aromatic monomer represented by  
 12 the following formula;



- 13  
 14 wherein  $R^1$  is selected from the group of radicals consisting  
 15 of hydrogen and alkyl radicals containing three carbons or  
 16 less, and Ar is a phenyl group or a phenyl group substituted  
 17 with from 1 to 5 substituents selected from the group  
 18 consisting of halo,  $C_{1-4}$ -alkyl, and  $C_{1-4}$ -haloalkyl; or
- 19 (b) said hindered aliphatic or cycloaliphatic vinyl or vinylidene  
 20 monomer represented by the following general formula;



- 21  
 22 wherein  $A^1$  is a sterically bulky, aliphatic or cycloaliphatic  
 23 substituent of up to 20 carbons,  $R^1$  is selected from the group  
 24 of radicals consisting of hydrogen and alkyl radicals  
 25 containing from 1 to 4 carbon atoms, preferably hydrogen or  
 26 methyl; each  $R^2$  is independently selected from the group of  
 27 radicals consisting of hydrogen and alkyl radicals containing

- 28 from 1 to 4 carbon atoms, preferably hydrogen or methyl; or  
29 alternatively R<sup>1</sup> and A<sup>1</sup> together form a ring system; or  
30 (c) a combination of at least one aromatic vinyl or vinylidene  
31 monomer and at least one hindered aliphatic or cycloaliphatic  
32 vinyl or vinylidene monomer, and  
33 (2) from 45 to 99 mol percent of polymer units derived from ethylene,  
34 or ethylene and at least one of propylene, 4-methyl-1-pentene,  
35 butene-1, hexene-1 or octene-1; and  
36 iii) wherein said tackifier, Component IB, when present, is present in an  
37 amount from 5 to 50 percent by weight (based on the combined weights of  
38 components IA and IB) and comprises a wood rosin, a tall oil derivative, a  
39 cyclopentadiene derivative, a natural terpene, a synthetic terpene, a terpene-  
40 phenolic resin, a styrene/ $\alpha$ -methyl styrene resin, or a mixed aliphatic-  
41 aromatic tackifying resin, or any combination thereof; and  
42 (iv) said second component, II, is present in amount of from 5 to 75 wt  
43 percent (based on the combined weights of Components I and II) which  
44 comprises one or more of;  
45 A) an ethylene or  $\alpha$ -olefin homopolymer or interpolymer;  
46 B) an ethylene/propylene rubber (EPM), ethylene/propylene diene  
47 monomer terpolymer (EPDM), isotactic polypropylene;  
48 C) a styrene/ethylene-butene copolymer, a styrene/ethylene-  
49 propylene copolymer, a styrene/ethylene-butene/styrene (SEBS)  
50 copolymer, a styrene/ethylene-propylene/styrene (SEPS)  
51 copolymer,  
52 D) the acrylonitrile-butadiene-styrene (ABS) polymers, styrene-  
53 acrylonitrile (SAN), high impact polystyrene,  
54 E) epoxies, vinyl ester resins, polyurethanes, phenolic resins,  
55 F) poly(methylmethacrylate), polyester, nylon-6, nylon-6,6, poly(acetal);  
56 poly(amide), poly(arylate), poly(carbonate), poly(butylene) and  
57 polybutylene, polyethylene terephthalates.

- 1 16. The bicomponent fiber of Claim 15 wherein;
- 2 (i) said first component I comprises from 50 to 95 wt percent (based on the
- 3 combined weights of Components I and II);
- 4 (ii) Component (IA) is present in an amount of 60 to 90 wt percent (based on
- 5 the combined weights of Components IA and IB) and comprises at least
- 6 one substantially random interpolmer having an  $I_2$  of 0.5 to 100 g/10 min,
- 7 a density of from 0.930 to 1.040 g/cm<sup>3</sup> and an  $M_w/M_n$  of 2 to 5; which
- 8 comprises;
- 9 (1) from 2 to 50 mol percent of polymer units derived from;
- 10 a) said vinyl or vinylidene aromatic monomer which comprises
- 11 styrene,  $\alpha$ -methyl styrene, ortho-, meta-, and para-
- 12 methylstyrene, and the ring halogenated styrenes, or
- 13 b) said hindered aliphatic or cycloaliphatic vinyl or vinylidene
- 14 monomers which comprises 5-ethylidene-2-norbornene or 1-
- 15 vinylcyclo-hexene, 3-vinylcyclo-hexene, and 4-
- 16 vinylcyclohexene;
- 17 (c) a combination of at least one aromatic vinyl or vinylidene
- 18 monomer and at least one hindered aliphatic or cycloaliphatic
- 19 vinyl or vinylidene monomer, and
- 20 (2) from 50 to 98 mol percent of polymer units derived from ethylene,
- 21 or ethylene and at least one of propylene, 4-methyl-1-pentene,
- 22 butene-1, hexene-1 or octene-1; and
- 23 iii) said tackifier, Component IB, when present is present in an amount from 10
- 24 to 40 percent by weight (based on the combined weights of components IA
- 25 and IB) and comprises a styrene/ $\alpha$ -methyl styrene resin, or a mixed
- 26 aliphatic-aromatic tackifying resin or any combination thereof; and
- 27 (iv) said second component, II, is present in amount of from 5 to 50 wt
- 28 percent (based on the combined weights of Components I and II) which
- 29 comprises one or more of;
- 30 A) an ethylene or  $\alpha$ -olefin homopolymer or interpolmer;

- 31 B) a styrene/ethylene-butene copolymer, a styrene/ethylene-  
32 propylene copolymer, a styrene/ethylene-butene/styrene (SEBS)  
33 copolymer, a styrene/ethylene-propylene/styrene (SEPS)  
34 copolymer, high impact polystyrene,  
35 C) poly(methylmethacrylate), polyester, nylon-6, nylon-6,6,  
36 poly(acetal); poly(amide), poly(arylate), poly(carbonate),  
37 poly(butylene) and polybutylene, polyethylene terephthalates.

1 17. The bicomponent fiber of Claim 16 wherein said fiber is of the core/sheath type  
2 and wherein Component I is the core and Component II is the sheath and wherein  
3 Component IA1 is styrene; and Component IA2 is ethylene; Component IB is not  
4 present and Component II is polypropylene, polyethylene, ethylene/octene  
5 copolymer, polyethylene terephthalate, polystyrene, nylon-6, nylon-6,6, or  
6 combinations thereof.

1 18. The bicomponent fiber of Claim 18 wherein said fiber is of the core/sheath type  
2 and wherein Component I is the core and Component II is the sheath and wherein  
3 Component IA1 is styrene; and Component IA2 is ethylene and at least one of  
4 propylene, 4-methyl-1-pentene, butene-1, hexene-1 or octene-1; Component IB is  
5 not present and Component II is polypropylene, polyethylene, ethylene/octene  
6 copolymer, polyethylene terephthalate, polystyrene, nylon-6, nylon-6,6, or  
7 combinations thereof.

8

1 19. A fabric comprising the fiber of Claim 14.

1 20. The fabric of Claim 19, comprising a woven fabric.

1 21. The fabric of Claim 19, comprising a non-woven fabric.

1 22. A fabricated article prepared from the fiber of Claim 14, comprising carpet, doll  
2 hair, a wig, a tampon, a diaper, athletic sportswear, wrinkle free and form-fitting  
3 apparel, upholstery, bandages, and gamma sterilizable non-woven articles.

1 23. A plurality of the fibers of Claim 14 in the form of doll hair.

1 24. A plurality of the fibers of Claim 17 in the form of doll hair.

1 25. A plurality of the fibers of Claim 18 in the form of doll hair.



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/05285

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 D01F6/30 D01F6/42 D01F6/46 D01F6/56 D01F8/06  
A63H3/44

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 D01F A63H

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	WO 98 27156 A (BETHEA JAMES R ;CHEUNG YUNWA W (US); DOW CHEMICAL CO (US); GATHERS) 25 June 1998 see page 13, line 16 - line 21; claims 1-12	1,6-13
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Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

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## INTERNATIONAL SEARCH REPORT

International Application No

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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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